# Classical and Quantum Nonlinear Optics

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# Contents

# Part 1: Classical Nonlinear Optics

# Chapter 1

# Nonlinear optical response

The subject we will take up for the majority of this course is what we call *nonlinear optics*, which concerns itself with the interaction of high intensity electromagnetic radiation with materials.

## 1.1 The fundamental idea: nonlinear response to light

Let us introduce nonlinear response by contrasting with the case of linear response. Suppose we apply an electric field to a material. All materials are polarizable to some extent: if an electric field is present inside them, then it will move charges around to induce polarization $\mathbf{P}$. In the linear electrodynamics of continuous media, we say that the polarization is linear in the electric field with some proportionality constant $\chi$. The simplest linear relationship that one could write is

$$\mathbf{P}(\mathbf{r}, t) = \epsilon_0 \chi \mathbf{E}(\mathbf{r}, t). \tag{1.1.1}$$

This is an *instantaneous* and *local* relationship between the induced polarization and the electric field: the induced polarization at position $\mathbf{r}$ and time $t$ depends on the electric field $\mathbf{E}$ at position $\mathbf{r}$ and time $t$. The electric field is the *total* electric field, which differs from the applied field insofar as the total field is the sum of the externally applied electric field and the electric field associated with the induced polarization.

The instantaneous, local relationship is not the most general linear relationship one could write. Indeed, if we think of the polarization as a linear functional of the electric field $\mathbf{P}[\mathbf{E}(\mathbf{r}, t)]$, then the most general linear relationship is one where the induced polarization $\mathbf{P}$ at $\mathbf{r}, t$ is a linear superposition of the electric field at all other positions and times [1].

$$\mathbf{P}(\mathbf{r}, t) = \epsilon_0 \int d\mathbf{r}_1 dt_1 \; \tilde{\chi}(\mathbf{r}, \mathbf{r}_1, t, t_1) \mathbf{E}(\mathbf{r}_1, t_1). \tag{1.1.2}$$

If we now ask: what is the most general functional $\mathbf{P}[\mathbf{E}]$, there is no reason why that relationship need be linear. We can however imagine that even if the relationship is *nonlinear*, for weak enough fields, we could approximate the relationship as linear. And indeed, that's

[1] You might object that the polarization cannot depend on spacetime points with spacelike separations, or even times *later* than the time in question. Both of these considerations work their way in as constraints on the linear susceptibility.

the basic idea of nonlinear optics. The invention of the laser gave us access to focusable electromagnetic radiation of significant intensity to see the influence of the nonlinear terms in the polarization. In this class, we will develop the consequences of a nonlinear relationship between the polarization and electric field.

If the electromagnetic field is strong, but not *so* strong, we expect that we can represent the polarization in terms of a low-order Taylor expansion in powers of the electric field. We express this Taylor series as [2]

$$\mathbf{P}(\mathbf{r},t) = \sum_{n=1}^{\infty} \mathbf{P}^{(n)}(\mathbf{r},t), \tag{1.1.3}$$

where

$$P_i^{(n)}(\mathbf{r},t) = \epsilon_0 \int \left( \prod_{i=1}^{n} d\mathbf{r}_i dt_i \right) \tilde{\chi}^{(n)}_{i,i_1,\cdots,i_n}(\mathbf{r},\mathbf{r}_1\cdots\mathbf{r}_n,t,t_1\cdots t_n) E_{i_1}(\mathbf{r}_1,t_1)\cdots E_{i_n}(\mathbf{r}_n,t_n). \tag{1.1.4}$$

In this expression, we have made use of repeated index notation: $T_{ij}a_ib_j = \sum_{i,j} T_{ij}a_ib_j$. We will generally make use of repeated index notation. This expression while appearing complicated, is simply a dressed up multivariate Taylor expansion where the variables are $E_i(\mathbf{r},t)$: the electric field of a given component, at a given position, and a given time is treated as a variable [3]. The quantity $\tilde{\chi}^{(n)}_{i,i_1,\cdots,i_n}(\mathbf{r},\mathbf{r}_1\cdots\mathbf{r}_n,t,t_1\cdots t_n)$ is called the $n$th order nonlinear susceptibility.

Although it is relatively straightforward to understand how a relationship like this comes about, it is still extremely complicated, and we will need to simplify this. In this chapter we will develop general constraints on the form of the nonlinear susceptibility, which will arm us, in Chapter 2, to consider specific nonlinear interactions. We will illustrate these ideas by computing the nonlinear susceptibility for a specific microscopic model: a classical anharmonic oscillator. This microscopic model will also give us some indication of what electric field strengths are needed to access nonlinearities. In this chapter, we will also see how the existence of these nonlinear responses allows for phenomena such as harmonic generation, sum-frequency generation, difference-frequency generation, and self-phase modulation.

## 1.2 Classical anharmonic oscillator as a microscopic model of nonlinear response

In this section, we will develop a simple concrete model of nonlinear optical response. This model will allow us to see: what is needed microscopically to get nonlinear response, what effects come from nonlinear response, why high-intensity is needed to access nonlinear effects, and many other general properties of nonlinear response tensors. This is a *very important section*: study it well, and you will have intuition about nonlinear optics *as a whole*.

[2] I omitted permament polarization, as in say a water molecule.

[3] Consider a scalar function $f(\mathbf{x})$ of $n$ variables $x_1\cdots x_n$. Then the $k$th term in the multivariate Taylor expansion, expanded about $\mathbf{x}=0$ is (in repeated index notation) $\frac{1}{k!}\frac{\partial^k f}{\partial x_{i_1}\cdots\partial x_{i_k}} x_{i_1}\cdots x_{i_k}$. The derivative term is essentially the nonlinear susceptibility and we have expressed only the vector index sums in repeated index notation while treating the "sum" over continuous variables as integrals.

### 1.2.1 Linear oscillator

Let us consider a rudimentary model of a polarizable material as a collection of independent and identical harmonic oscillators of density $n$, mass $m$, charge $q$, natural frequency $\omega_0$, and damping constant $\gamma$. If the material is overall neutral and so the picture is that each element (atom) of the material is composed of one stationary charge of magnitude $-q$ (representing for example the nucleus which is much more massive than the electrons) and one movable charge of charge $q$ (representing for example an electron). This model is called the *Lorentz oscillator*.

Each oscillator couples to the electric field $\mathbf{E}$. The equation of motion for the position $\mathbf{x}$ of each oscillator is

$$\ddot{\mathbf{x}} + \gamma\dot{\mathbf{x}} + \omega_0^2\mathbf{x} = \frac{q\mathbf{E}}{m}. \tag{1.2.1}$$

This equation is most readily solved in Fourier domain, and the solution can easily be seen to be

$$\mathbf{x}(t) = \int \frac{d\omega}{2\pi}\, e^{-i\omega t}\frac{q\mathbf{E}(\omega)/m}{D(\omega)}, \tag{1.2.2}$$

where $D(\omega) \equiv (\omega_0^2 - \omega^2) - i\gamma\omega$. Note that the frequency integration limits are $-\infty$ to $\infty$. The displacement of these charges constitutes a dipole. The polarization density is simply

$$\mathbf{P}(t) = nq\mathbf{x}(t) \leftrightarrow \mathbf{P}(\omega) = nq\mathbf{x}(\omega). \tag{1.2.3}$$

In frequency domain, we may then write that

$$\mathbf{P}(\omega) = \frac{nq^2}{mD(\omega)}\mathbf{E}(\omega), \tag{1.2.4}$$

which allows us to identify the linear susceptibility as

$$\chi^{(1)}(\omega) = \frac{nq^2}{m\epsilon_0 D(\omega)} = \frac{nq^2}{m\epsilon_0}\frac{1}{(\omega_0^2 - \omega^2) - i\gamma\omega}. \tag{1.2.5}$$

The permittivity is related to the susceptibility by $\epsilon = \epsilon_0(1 + \chi^{(1)})$ and the index of refraction by $n = \sqrt{\epsilon/\epsilon_0}$. I have neglected the tensorial nature of the permittivity since the response to all field directions is the same. Therefore as a tensor, we have $\chi^{(1)}_{ij} = \chi^{(1)}\delta_{ij}$. If the natural frequency and damping constants were *different* in different directions $\omega^2_{0,x} \neq \omega^2_{0,y} \neq \omega^2_{0,z}$ (where $x, y, z$ refer to three orthogonal principal directions), then we get $\chi_{ii}(\omega) = \frac{nq^2}{m\epsilon_0 D_i(\omega)}$, where $D_i(\omega) = (\omega^2_{0,i} - \omega^2) - i\gamma_i\omega$ (and $\chi_{ij} = 0$ if $i \neq j$).

Let us review the important limits of this Lorentz oscillator.

1. At low frequencies, $\omega \ll \gamma, \omega_0$ we have that the permittivity is *constant* and real, and given by $nq^2/m\epsilon_0$: this is the static permittivity. At frequencies $\omega_0 \gg \omega \gg \gamma$, one gets the same result. Thus, for frequencies much smaller than the resonance frequency of the system, the permittivity is approximately non-dispersive (frequency-independent) and real. At optical ($\omega_0 \sim 10^16$rad/s) frequencies and for densities typical in solids ($10^{29}m^{-3}$), one gets a susceptibility of order 1.

2. On resonance, the susceptibility becomes *purely imaginary* $\chi(\omega) = inq^2/(m\epsilon_0\gamma\omega_0)$ and large in magnitude (compared to one) when $\gamma \ll \omega_0$. The imaginary part of the susceptibility is related to the rate of absorption of energy by the oscillator, and so on resonance, the large imaginary part of the susceptibility signifies strong absorption.

3. At high frequencies, the susceptibility goes to zero: this is because for high frequencies, the oscillator cannot keep up with the drive. The oscillator can only respond to the field over a timescale $\omega_0^{-1}$ and so over that timescale the electric field looks like it flips sign many times, averaging out to nearly no driving force. This behavior, that the permittivity goes to zero at high frequencies, is universal to all materials.

### 1.2.2 Cubic oscillator

The solution to the Lorentz oscillator is *exact*, and the polarization is linear in the electric field. To get contributions which are nonlinear in the electric field, the potential has to be *anharmonic* (in other words, it has to be a non-quadratic potential). In this section, we consider the example of a *cubic anharmonic oscillator* in one dimension. The relevant equation of motion becomes

$$\ddot{x} + \gamma\dot{x} + \omega_0^2 x + max^2 = qE(t)/m. \tag{1.2.6}$$

This equation is not exactly soluble. We will solve it *perturbatively*, assuming that the linear Lorentz oscillator behavior dominates (we will check for self-consistency after solving this equation). We pursue a perturbation theory in powers of the electric field.

$$x(t) = \sum_{k=1}^{\infty} x_k[E^k](t), \tag{1.2.7}$$

where $x_k[E^k]$ is a functional which has $k$ powers of the electric field. The sum starts from $k = 1$ since no field implies no response if the system does not have permanent dipole moments. To find the $x_k$: one can plug in the series expansion and equate terms order-by-order. In the quadratic term in the equation of motion, the lowest power of the electric field is two, and does not contribute to the solution for $x_1$. Therefore

$$\ddot{x}_1 + \gamma\dot{x}_1 + \omega_0^2 x_1 = qE(t)/m. \tag{1.2.8}$$

The solution is simply the Lorentz oscillator solution:

$$x_1(t) = \int \frac{d\omega}{2\pi}\, e^{-i\omega t} \frac{qE(\omega)/m}{D(\omega)}. \tag{1.2.9}$$

The first correction to the Lorentz oscillator behavior satisfies $\ddot{x}_2 + \gamma\dot{x}_2 + \omega_0^2 x_2 + ax_1^2 = 0$ and thus:

$$\ddot{x}_2 + \gamma\dot{x}_2 + \omega_0^2 x_2 = \int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi} e^{-i(\omega_1+\omega_2)t} \frac{-a(q/m)^2 E(\omega_1)E(\omega_2)}{D(\omega_1)D(\omega_2)}. \tag{1.2.10}$$

This can be solved by Fourier transformation. Fourier transforming both sides of the equation yields

$$\left[(\omega_0^2 - \omega^2) - i\gamma\omega\right] x_2(\omega) = \int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi}\, 2\pi\delta(\omega - (\omega_1+\omega_2)) \times \frac{-a(q/m)^2 E(\omega_1)E(\omega_2)}{D(\omega_1)D(\omega_2)}, \tag{1.2.11}$$

with solution

$$x_2(\omega) = \int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi}\ 2\pi\delta(\omega-\omega_\sigma) \times \frac{-a(q/m)^2 E(\omega_1)E(\omega_2)}{D(\omega_\sigma)D(\omega_1)D(\omega_2)}, \tag{1.2.12}$$

where we have defined $\omega_\sigma = \omega_1 + \omega_2$. The corresponding nonlinear polarization is $P^{(2)}(\omega) = nqx_2(\omega)$. As advertised we have a nonlinear relationship between the polarization and the field, which should define a $\chi^{(2)}$. We can define the second-order nonlinear susceptibility as follows:

$$P^{(2)}(\omega) = \epsilon_0 \int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi}\ 2\pi\delta(\omega-\omega_\sigma)\chi^{(2)}(\omega_1,\omega_2))E(\omega_1)E(\omega_2), \tag{1.2.13}$$

where

$$\chi^{(2)}(\omega_1,\omega_2) = -\frac{anq^3}{m^2\epsilon_0} \times \frac{1}{D(\omega_\sigma)D(\omega_1)D(\omega_2)}. \tag{1.2.14}$$

This definition is introduced to be consistent with standard texts in nonlinear optics. As can be very clearly seen, a field that has Fourier components at frequencies $\omega_1$ and $\omega_2$ generates polarization with Fourier components as $\omega_1 \pm \omega_2$. Why minus? Because a real electric field always has its Fourier components come in $\pm\omega$ (a real field satisfies $E(\omega) = E(-\omega)^*$).

### Harmonic generation

One such consequence of this frequency-combining is second harmonic generation. If we have a field component at frequency $\omega_1$ then one term in the polarization expansion is $P(2\omega) \sim \chi^{(2)}(\omega,\omega)E(\omega)^2$. Recalling from Maxwell's equations that a time-oscillating polarization at frequency $\omega$ produces a time-varying electric field at the same frequency, we see that by illuminating a material with second-order nonlinearity with a field at frequency $\omega$, we can generate a new electric field at frequency $2\omega$. This is called second-harmonic generation and is how nearly all green lasers work. They are based on the frequency-doubling of a relatively intense infrared laser (often based on Nd:YAG).

### Sum-frequency and difference-frequency generation

Similarly, if we have a field with frequency components $\omega_1$ and $\omega_2$, we can generate a new field with frequency $\omega_1 + \omega_2$: second harmonic generation is a special case. This is called sum-frequency generation. Also interesting is that a field at frequency $\omega_1 - \omega_2$ can be generated. This is called difference-frequency generation, and is an important technique for generating terahertz radiation. Consider a short optical pulse with complex field $E_0 e^{-i\omega_0 t} e^{-t^2/2\tau^2}$. It's Fourier transform is proportional to $e^{-(\omega-\omega_0)^2\tau^2}$: two frequency components separated by $\Delta\omega$ can combine to give a field at frequency $\Delta$. As can be seen from the Fourier domain field, $\Delta$ can be arbitrarily close to zero, and so a terahertz field can be generated.

### Optical rectification and Pockels effect

One also notices immediately from the preceding discussion of difference-frequency generation that *zero* frequency radiation can be generated by a second-order nonlinear medium. This is called optical rectification. The inverse process also exists. Suppose we have a superposition of a DC field and an AC field at frequency $\omega$. Then there is a polarization that can be created

at $\omega$ which is proportional to $E_{\rm DC}E(\omega)$. This looks like a *linear* relationship between the AC field and the polarization, and indeed, we can say that the DC field is changing the linear susceptibility! This is the Pockels effect, and leads to the ability to shift the phase of an incident light field with DC voltages. By applying a slow AC voltage, it is also possible to modulate the phase and amplitude of a light wave in time!

We have already seen that the presence of an anharmonic potential leads to sum and difference frequency generation (including harmonic generation and rectification as special cases), as well as the modulation of the linear index of refraction. The generation of new frequencies, and the modification of the linear index of refraction with an external control field, are in a sense, the main effects of nonlinear optics.

### 1.2.3 Quartic oscillator

We now explore the consequences of a quartic term in the potential. One might argue that the quartic term will provide sub-leading corrections to the cubic term. But the cubic term doesn't exist in a wide class of *centrosymmetric* materials. If the material looks the same upon inversion: $\mathbf{x} \to -\mathbf{x}$, then we expect that reversing the sign of an applied electric field would reverse the sign of the polarization. Even-order terms in the polarization expansion are incompatible with this requirement, thus forcing that all even-order nonlinear susceptibilities must vanish. Thus, in a centrosymmetric material, the lowest-order nonlinear polarization is the cubic term.

Let us consider a centrosymmetric quartic oscillator. The resulting tensor properties of the third-order nonlinear susceptibility depend on the exact symmetries of the potential. We will consider an oversimplified model and consider an *isotropic* potential of the form $V(\mathbf{x}) = \frac{1}{4}mb(\mathbf{r}\cdot\mathbf{r})^2$. The resulting equation of motion is

$$\ddot{\mathbf{x}} + \gamma\dot{\mathbf{x}} + \omega_0^2\mathbf{x} + b(\mathbf{x}\cdot\mathbf{x})\mathbf{x} = q\mathbf{E}(t)/m. \tag{1.2.15}$$

Pursuing a perturbation series in powers of the field, it is clear that the part of the field at linear order in the field is simply the Lorentz oscillator solution:

$$\mathbf{x}(t) = \int \frac{d\omega}{2\pi}\, e^{-i\omega t}\frac{q\mathbf{E}(\omega)/m}{D(\omega)}. \tag{1.2.16}$$

There is no term at quadratic order in the fields: the lowest order term that the cubic force term can generate is cubic in the electric field. The third-order response follows immediately as

$$\mathbf{x}_3(t) = \frac{-bq^3}{m^3}\int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi}\frac{d\omega_3}{2\pi}e^{-i\omega_\sigma t}\frac{(\mathbf{E}(\omega_1)\cdot\mathbf{E}(\omega_2))\,\mathbf{E}(\omega_3)}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)}, \tag{1.2.17}$$

with $\omega_\sigma = \omega_1 + \omega_2 + \omega_3$. The resulting nonlinear polarization is then given by

$$\mathbf{P}^{(3)}(\omega) = \frac{-nbq^4}{m^3}\int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi}\frac{d\omega_3}{2\pi}2\pi\delta(\omega-\omega_\sigma)\frac{(\mathbf{E}(\omega_1)\cdot\mathbf{E}(\omega_2))\,\mathbf{E}(\omega_3)}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)} \tag{1.2.18}$$

In component notation, this is

$$P_i^{(3)}(\omega) = \frac{-nbq^4}{m^3}\int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi}\frac{d\omega_3}{2\pi}2\pi\delta(\omega-\omega_\sigma)\frac{(E_j(\omega_1)E_j(\omega_2))\,E_i(\omega_3)}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)}. \tag{1.2.19}$$

The corresponding third-order susceptibility follows as

$$\chi^{(3)}_{ijkl} = -\frac{nq^4}{m^3\epsilon_0}\frac{1}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)}\delta_{jk}\delta_{il} \tag{1.2.20}$$

In the expression for $P_i^{(3)}(\omega)$, it is very clear that we could have written it as

$$P_i^{(3)}(\omega) = \frac{-nbq^4}{m^3}\int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi}\frac{d\omega_3}{2\pi}2\pi\delta(\omega-\omega_\sigma)\frac{(E_j(\omega_1)E_j(\omega_3))\,E_i(\omega_2)}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)}, \tag{1.2.21}$$

in which case we might have written $\chi^{(3)}_{ijkl} = -\frac{nq^4}{m^3\epsilon_0}\frac{1}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)}\delta_{jl}\delta_{ik}$, or

$$P_i^{(3)}(\omega) = \frac{-nbq^4}{m^3}\int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi}\frac{d\omega_3}{2\pi}2\pi\delta(\omega-\omega_\sigma)\frac{(E_j(\omega_2)E_j(\omega_3))\,E_i(\omega_1)}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)}, \tag{1.2.22}$$

in which we would have written $\chi^{(3)}_{ijkl} = -\frac{nq^4}{m^3\epsilon_0}\frac{1}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)}\delta_{kl}\delta_{ij}$. These are of course all the same expression and so we instead elect (for reasons to be made clearer later) to write the third-order susceptibility in a manifestly permutation-invariant way. Thus, we settle on the expression

$$\chi^{(3)}_{ijkl} = -\frac{nq^4}{3m^3\epsilon_0}\frac{\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)}. \tag{1.2.23}$$

**Frequency generation**

Compared to the case of second-harmonic generation, we see now that we can generate fields at frequencies $\omega_1 \pm \omega_2 \pm \omega_3$. One consequence of this is third-harmonic generation. There is also sum and frequency difference generation, just now involving three frequencies.

**Self-phase modulation**

A unique effect to third-order nonlinearity is self-phase modulation. This is associated with $\omega_1 = \omega, \omega_2 = \omega, \omega_3 = -\omega$ and always occurs (recall that any AC field has its Fourier components come in $\pm\omega$ pairs due to the reality of the field). This corresponds to a polarization at the *same* frequency as the field and acts as if the linear susceptibility were intensity dependent.

**Cross-phase modulation**

If we take $\omega_1 = \omega', \omega_2 = \omega, \omega_3 = -\omega$ then we get a polarization at frequency $\omega'$. This corresponds to a change in the index of refraction at frequency $\omega'$ controlled by the intensity of the field at frequency $\omega'$.

### 1.2.4 Intensity scale for nonlinear response

We have assumed that the nonlinear polarization terms are small compared to the linear polarizations. Let us now estimate the electric fields required to violate this assumption. To make matters simple, we will consider what happens at low frequencies. Let's consider the second-order susceptibility. The main problem is that we don't know what $a$ should be yet. We can find the approximate magnitude of $a$ as follows.

## 1.3 Properties of the nonlinear susceptibility

In this section, we work out a few key properties of the nonlinear susceptibility.

### 1.3.1 Locality

The relationship we wrote, (1.1.4) looks different than what is typically written in that we have allowed for a generally *nonlocal* response: the polarization at $\mathbf{r}$ could depend on the field at different points $\mathbf{r}'$. That is typically negligible in nonlinear optics constants as it is only important when the electric field actually *varies* on a scale comparable to the scale of variation of $\tilde{\chi}(\mathbf{r}, \mathbf{r}')$ (I have only retained two arguments here for clarity). That scale is the atomic scale, and since optical fields have negligible variations on this scale, the $\mathbf{r}'$ dependence is always integrated over in a way that has no dependence on the electric fields (since they can be pulled out of the spatial integrals). This leaves us with a simpler, spatially local relation:

$$P_i^{(n)}(\mathbf{r}, t) = \epsilon_0 \int \left( \prod_{i=1}^{n} dt_i \right) \tilde{\chi}^{(n)}_{i,i_1,\cdots,i_n}(\mathbf{r}, t, t_1 \cdots t_n) E_{i_1}(\mathbf{r}, t_1) \cdots E_{i_n}(\mathbf{r}, t_n). \tag{1.3.1}$$

Note that in writing this, I have not necessarily assumed that the nonlinear response is the same at every point in space: as might be the case if I had a composite material. In other words, I do not necessarily take *space-translation invariance to be a given.* In a uniform bulk material, the $\mathbf{r}$ dependence would go away.

### 1.3.2 Time-translation invariance

Suppose we subject the system to a pulsed electric field $E(\mathbf{r}, t)$[4] that arrives in some material at time $t = 0$. That is going to initiate polarization dynamics $P(\mathbf{r}, t)$. Now suppose I shift this electric field in time so that the pulse arrives in the material at a time $t = t_0$. In other words: $E(\mathbf{r}, t) \to E(\mathbf{r}, t - t_0)$. The statement of time-translation invariance is that the resulting polarization response is *also* shifted in time: $P(\mathbf{r}, t) \to P(\mathbf{r}, t - t_0)$. If we do not explicitly modify our material in time (or modify it quickly enough), time-translation invariance can be taken to hold.

Time-translation invariance is a powerful constraint. Suppose we shift our electric field by time $t_0$, then we can write the resulting polarization as [5]:

$$P_i^{(n)}(\mathbf{r}, t) = \epsilon_0 \int \left( \prod_{i=1}^{n} dt_i \right) \tilde{\chi}^{(n)}_{i,i_1,\cdots,i_n}(\mathbf{r}, t, t_1 \cdots t_n) E_{i_1}(\mathbf{r}, t_1 - t_0) \cdots E_{i_n}(\mathbf{r}, t_n - t_0). \tag{1.3.2}$$

[4] In this text, I will generally omit degrees of freedom if the relation holds for all possible values of the omitted degrees of freedom. In otherwords, if the relationship is an element-wise relationship. For example here, I have omitted the vector indices because time-translation symmetry holds for each vector component. We'll refer to this practice as omission of trivial labels. We will not always do this, and so if you think a label is missing, ask if the relation holds for all values of that label!

[5] You may wonder below why, in keeping with the practice of omitting trivial labels, I did not omit $\mathbf{r}$. This dependence is too important to omit!

However, this must be equal to the polarization induced by the *undelayed* fields up to a delay. In other words, the left-hand side must also be equal to:

$$\epsilon_0 \int \left( \prod_{i=1}^{n} dt_i \right) \tilde{\chi}^{(n)}_{i,i_1,\cdots,i_n}(\mathbf{r}, t - t_0, t_1 \cdots t_n) E_{i_1}(\mathbf{r}, t_1) \cdots E_{i_n}(\mathbf{r}, t_n). \tag{1.3.3}$$

Taking $t \to t - t_0$ in the above equation implies that

$$\tilde{\chi}^{(n)}(t, t_1 \cdots, t_n) = \tilde{\chi}^{(n)}(t - t_0, t_1 - t_0 \cdots, t_n - t_0), \tag{1.3.4}$$

where I have followed our practice of omitting the trivial space and direction labels. The nonlinear susceptibility is fully time-translation invariant. Let's go back to our relation for the nonlinear polarization and express it as:

$$P^{(n)}_i(\mathbf{r}, t) = \epsilon_0 \int \left( \prod_{i=1}^{n} dt_i \right) \tilde{\chi}^{(n)}_{i,i_1,\cdots,i_n}(\mathbf{r}, 0, t_1 - t \cdots t_n - t) E_{i_1}(\mathbf{r}, t_1) \cdots E_{i_n}(\mathbf{r}, t_n). \tag{1.3.5}$$

We may always perform this shift. We may then define a function of only $n$ time arguments: $\tilde{\chi}^{(n)}_{i,i_1,\cdots,i_n}(0, t_1 - t \cdots t_n - t) \to \chi^{(n)}_{i,i_1,\cdots,i_n}(t - t_1, \cdots, t - t_n)$, and define the nonlinear polarization at $n$th order by

$$P^{(n)}_i(\mathbf{r}, t) = \epsilon_0 \int \left( \prod_{i=1}^{n} dt_i \right) \chi^{(n)}_{i,i_1,\cdots,i_n}(\mathbf{r}, t - t_1, \cdots, t - t_n) E_{i_1}(\mathbf{r}, t_1) \cdots E_{i_n}(\mathbf{r}, t_n). \tag{1.3.6}$$

We will henceforth *always* write the susceptibility in this way and so we drop the tilde that we have been carrying around.

### 1.3.3 Frequency-domain representation of the susceptibility

The multi-convolutional form of the nonlinear polarization in (1.3.6) suggests a simpler relationship in frequency-domain. Frequency-domain is of course very important in optics because we often use light sources which are relatively narrow-band: in other words, they have a reasonably well-defined center frequency (this is even true of 100 femtosecond pulses). We may express the electric field in Fourier domain via

$$\mathbf{E}(\mathbf{r}, t) = \int \frac{d\omega}{2\pi}\, e^{-i\omega t} \mathbf{E}(\mathbf{r}, \omega) \leftrightarrow \mathbf{E}(\mathbf{r}, \omega) = \int dt\, e^{i\omega t} \mathbf{E}(\mathbf{r}, t). \tag{1.3.7}$$

Plugging this into (1.3.6) yields

$$P^{(n)}_i(\mathbf{r}, t) = \epsilon_0 \int \left( \prod_{i=1}^{n} dt_i \frac{d\omega_i}{2\pi} e^{-i\omega_i t_i} \right) \chi^{(n)}_{i,i_1,\cdots,i_n}(\mathbf{r}, t - t_1, \cdots, t - t_n) E_{i_1}(\mathbf{r}, \omega_i) \cdots E_{i_n}(\mathbf{r}, \omega_n), \tag{1.3.8}$$

which may be written as

$$P^{(n)}_i(\mathbf{r}, t) = \epsilon_0 \int \left( \prod_{i=1}^{n} \frac{d\omega_i}{2\pi} \right) e^{-i\omega_\sigma t} \chi^{(n)}_{i,i_1,\cdots,i_n}(\mathbf{r}, \omega_1, \cdots, \omega_n) E_{i_1}(\mathbf{r}, \omega_i) \cdots E_{i_n}(\mathbf{r}, \omega_n), \tag{1.3.9}$$

where $\omega_\sigma = \sum_{i=1}^{n} \omega_i$. In Fourier domain, the nonlinear polarization may be written as

$$P_i^{(n)}(\mathbf{r},\omega) = \epsilon_0 \int \left( \prod_{i=1}^{n} \frac{d\omega_i}{2\pi} \right) 2\pi\delta(\omega - \omega_\sigma) \chi^{(n)}_{i,i_1,\cdots,i_n}(\mathbf{r},\omega_1,\cdots,\omega_n) E_{i_1}(\mathbf{r},\omega_i) \cdots E_{i_n}(\mathbf{r},\omega_n). \tag{1.3.10}$$

The intuitive content of this equation is that an $n$th order nonlinear response converts $n$ fields at respective frequencies $\omega_1 \cdots \omega_n$ to a polarization at frequency $\omega_\sigma = \omega_1 + \cdots + \omega_n$.

**A brief note on notation**

The notation we are using here is not quite in keeping with Boyd and many other texts on nonlinear optics. For one, Boyd for example write frequency sums as discrete sums. This, in the instructor's view, is a very continuous-wave oriented picture where we think of light as consisting of a discrete sum of monochromatic frequencies. Of course, the discrete sum notation is more general than that! If we assume that our fields and nonlinear susceptibilities vary negligibly over some small frequency increment $\Delta\omega$, then we may write the Fourier integral as a Riemann sum. For a true sum of monochromatic waves, such that the field $\mathbf{E}(\omega) = \sum_n \delta(\omega - \omega_n)\mathbf{E}_n$, one could reduce the frequency integrations to sums weighted by the nonlinear susceptibilities evaluated at definite frequencies (being careful about regularizing the equations when two or more frequencies are identical).

Another notational departure is in the susceptibility. Almost all authors choose to write $\chi^{(n)}(\omega_1,\cdots,\omega_n)$ as $\chi^{(n)}(\omega_\sigma,\omega_1,\cdots,\omega_n)$ where one additional frequency argument is added at the beginning of the list of frequency arguments. This frequency is always $\omega_\sigma$. This argument is strictly speaking redundant but is a useful reminder as to what polarization is being generated! I will usually *not* write it unless I want to make explicit connection to canonical texts in nonlinear optics (e.g., Boyd).

### 1.3.4 Reality of the fields

The fact that the fields are real and the polarization is real requires that the time-domain susceptibility is real:

$$\chi^{(n)}(\tau_1 \cdots \tau_n) = (\chi^{(n)}(\tau_1 \cdots \tau_n))^*. \tag{1.3.11}$$

This immediately implies upon taking the Fourier transform of both sides that

$$\chi^{(n)}(-\omega_1 \cdots - \omega_n) = (\chi^{(n)}(\omega_1 \cdots \omega_n))^*. \tag{1.3.12}$$

### 1.3.5 Intrinsic permutation symmetry

In Eq. (1.3.10), it is clear that $\omega_1 \cdots \omega_n$ and $i_1 \cdots i_n$ are dummy variables that get summed over [6]. Suppose we define "super-indices" $\alpha_k = (i_k, \omega_k)$ and write the integral in Eq. (1.3.10) as a sum, leading to the form:

$$P_i^{(n)}(\omega) = \epsilon_0 \sum_{\alpha_1 \cdots \alpha_n} 2\pi\delta(\omega - \omega_\sigma) \chi^{(n)}_{i,\alpha_1 \cdots \alpha_n} E_{\alpha_1} \cdots E_{\alpha_n}. \tag{1.3.13}$$

[6] I will refer to discrete sums and integrals both as sums when appropriate and indices and continuous variables both as variables when appropriate.

For any given set of distinct indices, there are $\mathcal{N}_p$ distinct permutations of them which are all summed over. Denote the sum over permutations for a *fixed* set of indices, $\alpha_1 \cdots \alpha_n$ as $\sum_{\mathrm{P}[\alpha_1 \cdots \alpha_n]}$. Then

$$\sum_{\mathrm{P}[\alpha_1 \cdots \alpha_n]} = E_{\alpha_1} \cdots E_{\alpha_n} \sum_{\mathrm{P}[\alpha_1 \cdots \alpha_n]} \chi^{(n)}_{i,P[\alpha_1 \cdots \alpha_n]}. \tag{1.3.14}$$

We see that the susceptibility for any set of indices, is summed over all permutations of those indices. The resulting sum over permutations is clearly permutation symmetric. As a result, we are free to *define* the nonlinear susceptibility as being permutation symmetric with respect to the superindices (we cannot permute only the frequency indices, or only the spatial indices). We note that this is strictly speaking a choice, but it is a *harmless* choice because even if we defined the susceptibility to not have this permutation symmetry with respect to the superindices, we would only ever see its fully permutation-symmetric part [7]. Therefore, we *define* the susceptibility tensor to have the property

$$\chi^{(n)}_{i,P[\alpha_1 \cdots \alpha_n]} = \chi^{(n)}_{i,\alpha_1 \cdots \alpha_n}. \tag{1.3.15}$$

This property is called *intrinsic* permutation symmetry.

### 1.3.6 Causality and Kramers-Kronig relations

We have been taking the time integrals to run from $-\infty$ to $\infty$ implicitly. Of course, the response cannot precede the applied field. That immediately implies that the susceptibilities have Heaviside theta functions $\theta(t - t_i)$ inside of them for all time arguments. Using the property that $\theta(t - t_i)^2 = \theta(t - t_i)$, we may write for example:

$$\chi^{(n)}(\mathbf{r}, t - t_1, \cdots, t - t_n) = \chi^{(n)}(\mathbf{r}, t - t_1, \cdots, t - t_n)\theta(t - t_i) \text{ for any } i. \tag{1.3.16}$$

We can in fact multiply by any number of Heaviside functions of any time argument, so in general one could write

$$\chi^{(n)}(\mathbf{r}, t - t_1, \cdots, t - t_n) = \chi^{(n)}(\mathbf{r}, t - t_1, \cdots, t - t_n) \prod_{i=1}^{n} \theta(t - t_i)^{k_i}, k_i \in \mathbb{N}. \tag{1.3.17}$$

This relation implies a constraint between the real and imaginary parts of the Fourier transform of the response tensor, called the Kramers-Kronig relation. The relation follows from considering the Fourier representation of the Heaviside function,

$$\theta(t) = -\lim_{\eta \to 0} \int \frac{d\omega}{2\pi i} \frac{e^{-i\omega t}}{\omega + i\eta}. \tag{1.3.18}$$

and plugging it into (1.3.16). Fourier transforming both sides of the equation yields

$$\chi^{(n)}(\omega_1, \cdots, \omega_n) = -\lim_{\eta \to 0} \int \frac{d\omega}{2\pi i} \frac{\chi^{(n)}(\{\omega \neq \omega_i\}, \omega_i - \omega)}{\omega + i\eta}. \tag{1.3.19}$$

[7] The fully permutation symmetrized part of the tensor is $(\mathcal{N}_p!)^{-1} \sum_{\mathrm{P}[\alpha_1 \cdots \alpha_n]} \chi^{(n)}_{i,P[\alpha_1 \cdots \alpha_n]}$

Shifting variables leads to

$$\chi^{(n)}(\omega_1,\cdots,\omega_n) = -\lim_{\eta\to 0}\int \frac{d\omega}{2\pi i}\frac{\chi^{(n)}(\{\omega\neq\omega_i\},\omega)}{\omega_i-\omega+i\eta}. \tag{1.3.20}$$

To proceed, we need to make use of the distributional identity known as the Plemelj (or Sohotsky-Plemelj formula):

$$\lim_{\eta\to 0}\frac{1}{x-i\eta} = \mathcal{P}\left[\frac{1}{x}\right] + i\pi\delta(x), \tag{1.3.21}$$

where $\mathcal{P}$ denotes Cauchy principal value [8]. Using this identity, one arrives at

$$\chi^{(n)}(\omega_1,\cdots,\omega_n) = \frac{1}{i\pi}\mathcal{P}\int d\omega\frac{\chi^{(n)}(\{\omega\neq\omega_i\},\omega)}{\omega-\omega_i}, \tag{1.3.22}$$

which is the statement of the Kramers-Kronig relation.

The Kramers-Kronig relation is most commonly stated as a relationship between the real and imaginary parts of the function. For example, taking the real part of (1.3.22), we have

$$\text{Re }\chi^{(n)}(\omega_1,\cdots,\omega_n) = \frac{1}{\pi}\mathcal{P}\int d\omega\frac{\text{Im }\chi^{(n)}(\{\omega\neq\omega_i\},\omega)}{\omega-\omega_i}, \tag{1.3.23}$$

and

$$\text{Im }\chi^{(n)}(\omega_1,\cdots,\omega_n) = -\frac{1}{\pi}\mathcal{P}\int d\omega\frac{\text{Re }\chi^{(n)}(\{\omega\neq\omega_i\},\omega)}{\omega-\omega_i}, \tag{1.3.24}$$

**Linear case**

The by-far most useful application of Kramers-Kronig relations in optics is for *linear susceptibilities*. Although this is a course on nonlinear optics, this is a very important result and is often not covered in intermediate-level electrodynamics courses, so we discuss it here. In the linear case, we have

$$\text{Re }\chi^{(1)}(\omega) = \frac{1}{\pi}\mathcal{P}\int d\omega\frac{\text{Im }\chi^{(1)}(\omega')}{\omega'-\omega}, \tag{1.3.25}$$

and

$$\text{Im }\chi^{(1)}(\omega) = -\frac{1}{\pi}\mathcal{P}\int d\omega\frac{\text{Re }\chi^{(1)}(\omega')}{\omega'-\omega}. \tag{1.3.26}$$

The reality property for the linear susceptibility $\chi^{(1)}(\omega) = (\chi^{(1)}(-\omega))^*$ implies that the real part is even under frequency-domain reflection and the imaginary part is odd. We may use that to write the frequency integrals as ones with integration limits $0,\infty$ as

$$\text{Re }\chi^{(1)}(\omega) = \frac{2}{\pi}\mathcal{P}\int_0^\infty d\omega'\frac{\omega'\text{Im }\chi^{(1)}(\omega')}{\omega'^2-\omega^2}. \tag{1.3.27}$$

[8] The Cauchy principal value is a distribution defined such that $\mathcal{P}\int dx\frac{f(x)}{x-a} = \lim_{\epsilon\to 0}\left[\int^{a-\epsilon} + \int_{a+\epsilon}\right] f(x)$. In other words we integrate "everywhere except a".

For the imaginary part, we have

$$\text{Im } \chi^{(1)}(\omega) = -\frac{2\omega}{\pi}\mathcal{P}\int_0^\infty d\omega' \frac{\text{Re } \chi^{(1)}(\omega')}{\omega'^2 - \omega^2}. \tag{1.3.28}$$

A famous consequence of these relationships is that if the real part of the susceptibility (related to the index of refraction) is frequency-independent *everywhere* then the imaginary part of the susceptibility is zero identically. One may quickly check this from (1.3.27).

## 1.4 Dispersionless nonlinear media

In general, the nonlinear susceptibility depends on all of its frequency arguments. For materials where the polarization approximately responds instantaneously to the field, the susceptibility is *independent* of frequency. This is an approximation: no system responds instantaneously to any field, but if one has a system where the lowest resonance frequency is much larger than any of the frequencies in the electric field, the system can respond approximately instantaneously. We can see this in our model of the anharmonic oscillator in which for all $\omega \ll \omega_0$.

Then $\chi(\tau_1, \cdots, \tau_n) = \epsilon_0 \chi \delta(\tau_1) \cdots \delta(\tau_n)$ and

$$P_i(t) = \epsilon_0 \chi_{ii_1\cdots i_n} E_{i_1}(t) \cdots E_{i_n}(t). \tag{1.4.1}$$

It is also clear that

$$\chi(\omega_1 \cdots \omega_n) = \chi, \tag{1.4.2}$$

and so in frequency-domain, we have:

$$P_i^{(n)}(\mathbf{r}, \omega) = \epsilon_0 \int \left(\prod_{i=1}^n \frac{d\omega_i}{2\pi}\right) 2\pi\delta(\omega - \omega_\sigma)\chi_{ii_1\cdots i_n}(\mathbf{r}) E_{i_1}(\mathbf{r}, \omega_i) \cdots E_{i_n}(\mathbf{r}, \omega_n). \tag{1.4.3}$$

It is clear from the expressions above that the frequency-domain susceptibility for a dispersionless medium must be *real*.

Dispersionless nonlinear media have important properties.

### 1.4.1 Existence of a conserved energy

A dispersionless nonlinear medium has a conserved energy. We will find that energy here. We follow a different method from Boyd which is entirely classical [9]. A number of the results we derive here are derived using quantum mechanics in Boyd.

To arrive at a conserved energy, we are going to explicitly derive a Hamiltonian for the electromagnetic field. This is done by finding a Lagrangian that reproduces the equations of motion (the Maxwell equations with nonlinear polarization) and derive the corresponding

[9] In his approach, he appeals to a quantum mechanical derivation of the nonlinear susceptibilities to show various properties of a lossless / dispersionless medium. Our opinion is that one can arrive that these properties consistently classically, and so we "prefer" our approach though it requires additional apparatus!

Hamiltonian. We will use these results again later on. The Lagrangian which reproduces the *vacuum* Maxwell equations is (see Appendix B)

$$L = \int d\mathbf{r}\ \mathcal{L},\ \text{with}\ \mathcal{L} = \frac{\epsilon_0}{2}(\partial_t \mathcal{A})^2 - c^2(\nabla \times \mathbf{A})^2. \tag{1.4.4}$$

The Maxwell equations are arrived at through the Euler-Lagrange equation, which is

$$\frac{\partial \mathcal{L}}{\partial \phi} - \nabla_{\mathbf{r}} \cdot \frac{\partial \mathcal{L}}{\partial(\nabla_{\mathbf{r}}\phi)} = \partial_t \frac{\partial \mathcal{L}}{\partial(\partial_t \phi)}, \tag{1.4.5}$$

where $\phi$ refers to any component of the vector potential. For the vacuum Lagrangian, the corresponding Maxwell equation is

$$\nabla \times \nabla \times \mathbf{A} + \frac{1}{c^2}\partial_t^2 \mathbf{A} = 0, \tag{1.4.6}$$

which reduces to the correct Maxwell equation in the absence of free charges by noting that $\mathbf{E} = -\partial_t \mathbf{A}$.

In the presence of nonlinear sources, the equation of motion becomes

$$\nabla \times \nabla \times \mathbf{A} + \frac{1}{c^2}\partial_t^2 \mathbf{A} = \mu_0 \mathbf{J} = \mu_0 \partial_t \mathbf{P}[-\partial_t \mathbf{A}]. \tag{1.4.7}$$

We want to find a term of the Lagrangian density $\mathcal{L}_{\text{pol}}$ that produces this additional term in the equation of motion. It is not hard to guess that the term is

$$\mathcal{L}_{\text{pol}} = \epsilon_0 \sum_{n=1}^{\infty} \frac{1}{n+1} \chi^{(n)}_{i_0 i_1 i_2 \cdots i_n} E_{i_0} E_{i_1} \cdots E_{i_n} = \epsilon_0 \sum_{n=1}^{\infty} \frac{1}{n+1} \chi^{(n)}_{i_0 i_1 i_2 \cdots i_n} (\partial_t A_{i_0})(\partial_t A_{i_1}) \cdots (\partial_t A_{i_n}). \tag{1.4.8}$$

This Lagrangian density contributes to the equation of motion via a term

$$\frac{\partial}{\partial(\partial_t A_i)} \mathcal{L}_{\text{pol}} = \epsilon_0 \sum_{n=1}^{\infty} \chi^{(n)}_{i i_1 i_2 \cdots i_n} E_{i_0} E_{i_1} \cdots E_{i_n} = P. \tag{1.4.9}$$

In writing this, I have used the fact that the nonlinear Lagrangian density implicitly picks out the *fully symmetric* part of the susceptibility tensor. Since the Lagrangian generates dynamics, it must be the case that we can simply take the nonlinear susceptibility to be symmetric with respect to all of its indices. This is equivalent to a Kleinman symmetry [10].The resulting equation of motion is what we expect. The total Lagrangian density, including the parts that reproduce the vacuum-field Maxwell equations is

$$\mathcal{L} = \frac{\epsilon_0}{2}(\partial_t \mathcal{A})^2 - c^2(\nabla \times \mathbf{A})^2 + \mathcal{L}_{\text{pol}}. \tag{1.4.10}$$

The Lagrangian can be converted to a Hamiltonian density via

$$\mathcal{H} = \frac{\partial \mathcal{L}}{\partial(\partial_t \mathbf{A})} \cdot (\partial_t \mathbf{A}) - \mathcal{L} = \frac{\epsilon_0}{2}\left(\mathbf{E}^2 + c^2 \mathbf{B}^2\right) + \sum_{n=1}^{\infty} \frac{n}{n+1} \chi^{(n)}_{i_0 i_1 i_2 \cdots i_n} E_{i_0} E_{i_1} \cdots E_{i_n}. \tag{1.4.11}$$

[10] Note that the $n = 1$ term is the linear response, which can be included here without loss of generality.

For a system with time-translation symmetry, like this one, there is a conserved energy which is simply the integral of the Hamiltonian density. We may therefore say that the conserved energy is

$$U = \int d\mathbf{r}\mathcal{H} = \int d\mathbf{r}\ \left( \frac{\epsilon_0}{2} \left( \epsilon \mathbf{E}^2 + c^2 \mathbf{B}^2 \right) + \sum_{n=2}^{\infty} \frac{n}{n+1} \chi^{(n)}_{i_0 i_1 i_2 \cdots i_n} E_{i_0} E_{i_1} \cdots E_{i_n} \right), \tag{1.4.12}$$

where we have separated out the quadratic term in $\mathcal{L}_{\text{pol}}$ in order to separate the Lagrangian density into quadratic and non-quadratic parts. Occasionally, we will use a simplified notation for the nonlinear terms representing the full contraction of the nonlinear susceptibility against $(n+1)$ fields:

$$U = \int d\mathbf{r}\mathcal{H} = \int d\mathbf{r}\ \left( \frac{\epsilon_0}{2} \left( \epsilon \mathbf{E}^2 + c^2 \mathbf{B}^2 \right) + \sum_{n=2}^{\infty} \frac{n}{n+1} \chi^{(n)} : \mathbf{E}^{\otimes (n+1)} \right). \tag{1.4.13}$$

Since the energy associated with the electromagnetic field is conserved, we call such a system *lossless*.

### 1.4.2 Kleinman symmetry

By construction of a Lagrangian $\mathcal{L}$ that reproduces the Maxwell equations, it is required by construction that the nonlinear susceptibility can be taken to be *fully* permutation symmetric with respect to its indices. This is referred to Kleinman symmetry. There is an *related* symmetry called *full permutation symmetry* which says that we can permute *any* the indices (including the first) as long as we simultaneously interchange the frequency indices. Kleinman symmetry can be seen as a special case of full permutation symmetry in which the nonlinear susceptibility is frequency-independent.

#### Application to second-order susceptibility

The Kleinman symmetry can be used to represent second-order nonlinear polarization in a simplified manner. Define (as has been historically done)

$$d_{ijk} = \frac{1}{2} \chi^{(2)}_{ijk}. \tag{1.4.14}$$

In a lossless medium, the indices $d_{ijk}$ is completely symmetric under any interchange of indices: it is therefore symmetric with respect to interchange of $j$ and $k$. As a result, we can take $d_{ijk} \to d_{il}$ where $\ell = 1, ..., 6$ and maps to $(j,k)$ via:

$$(1,1) \to 1, (2,2) \to 2, (3,3) \to 3, (2,3)/(3,2) \to 4, (1,3)/(3,1) \to 5, (1,2)/(2,1) \to 6. \tag{1.4.15}$$

This reduced form is called *contracted notation*. Of course, we know that $d_{ijk}$ has *full* permutation symmetry, and so these 18 components of $d_{il}$ are not independent. There are

10 independent components [11]. The independent components can easily be found to be: $11, 12(=26), 13(=35), 14(=25=36), 15(=31), 16(=21), 22, 23(=34), 24(=32)$ and 33.

Using contracted notation (replacing $\chi_{ijk}$ by $d_{il}$), we may express second-order nonlinear phenomena more compactly. For example, consider sum-frequency generation of two monochromatic waves. Assuming that we have a field [12]

$$\mathbf{E}(t) = \mathbf{E}(\omega_1)e^{-i\omega_1 t} + \mathbf{E}(\omega_2)e^{-i\omega_2 t} + \mathbf{E}(\omega_1)^* e^{i\omega_1 t} + \mathbf{E}(\omega_2)^* e^{i\omega_2 t}. \tag{1.4.16}$$

The second-order polarization component at the sum-frequency is

$$P_i^{(2)}(\omega_1+\omega_2) = 2\epsilon_0 \chi_{ijk}^{(2)} E_{1,j} E_{2,k} = 4\epsilon_0 d_{ijk}^{(2)} E_j(\omega_1) E_k(\omega_2). \tag{1.4.17}$$

Written in terms of contracted notation, this could be written as

$$\begin{pmatrix} P_x^{(2)}(\omega_1+\omega_2) \\ P_y^{(2)}(\omega_1+\omega_2) \\ P_z^{(2)}(\omega_1+\omega_2) \end{pmatrix} = 4\epsilon_0 \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix} \begin{pmatrix} E_x(\omega_1)E_x(\omega_2) \\ E_y(\omega_1)E_y(\omega_2) \\ E_z(\omega_1)E_z(\omega_2) \\ E_y(\omega_1)E_z(\omega_2) + E_z(\omega_1)E_y(\omega_2) \\ E_x(\omega_1)E_z(\omega_2) + E_z(\omega_1)E_x(\omega_2) \\ E_x(\omega_1)E_y(\omega_2) + E_y(\omega_1)E_x(\omega_2) \end{pmatrix}. \tag{1.4.18}$$

Written in terms of independent components of the contracted $d$ "tensor", we could write:

$$\begin{pmatrix} P_x^{(2)}(\omega_1+\omega_2) \\ P_y^{(2)}(\omega_1+\omega_2) \\ P_z^{(2)}(\omega_1+\omega_2) \end{pmatrix} = 4\epsilon_0 \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{pmatrix} \begin{pmatrix} E_x(\omega_1)E_x(\omega_2) \\ E_y(\omega_1)E_y(\omega_2) \\ E_z(\omega_1)E_z(\omega_2) \\ E_y(\omega_1)E_z(\omega_2) + E_z(\omega_1)E_y(\omega_2) \\ E_x(\omega_1)E_z(\omega_2) + E_z(\omega_1)E_x(\omega_2) \\ E_x(\omega_1)E_y(\omega_2) + E_y(\omega_1)E_x(\omega_2) \end{pmatrix}. \tag{1.4.19}$$

For second-harmonic generation, it is relatively straightforward to see that

$$\begin{pmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{pmatrix} = 2\epsilon_0 \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{pmatrix} \begin{pmatrix} E_x^2(\omega) \\ E_y^2(\omega) \\ E_z^2(\omega) \\ 2E_y(\omega)E_z(\omega) \\ 2E_x(\omega)E_z(\omega) \\ 2E_x(\omega)E_y(\omega) \end{pmatrix}. \tag{1.4.20}$$

[11] Of the components of $\chi_{ijk}^{(2)}$ where all three indices are different, five of those six are redundant. Of the components where two indices are different (there are 18 such), 12 are redundant. Of components where all indices are the same, there are no such redundancies, leaving 10 out of 27 non-redundant components. Of course, it *must* be the case that $d_{il}$ carries the same information as $\chi_{ijk}^{(2)}$ and so the number of independent quantities in either must be the same.

[12] Your instructor would have preferred to multiply this expression by 1/2 but Boyd uses the convention below. To make the resulting expressions directly comparable to the textbook, I use (begrudgingly) the convention without the 1/2.

### 1.4.3 Effective value of d: $d_{\mathbf{eff}}$

It is sometimes useful to recast the relations for field and polarization as scalar relations. For example, it is possible to write for sum-frequency generation

$$P(\omega_1 + \omega_2) = 4\epsilon_0 d_{\text{eff}} E(\omega_1) E(\omega_2), \tag{1.4.21}$$

where $P = |\mathbf{P}|, E = |\mathbf{E}|$. The effective $d$ value depends on the exact symmetries of $d_{il}$. For example, as Boyd describes, for the $3m$ crystal class,

$$d_{\text{eff}} = d_{31} \sin\theta - d_{22} \cos\theta \sin(3\phi), \tag{1.4.22}$$

when the two lower-frequency waves have the same polarization, where $\theta$ is the angle between the propagation direction and the crystal axis (defined to be the $z$ direction). $\phi$ is the angle between the propagation vector and the $xz$ crystalline plane.

# Chapter 2

# Effects resulting from second-order optical nonlinearity

In the previous chapter, we developed the consequences of a nonlinear relationship between polarization and electric field, anticipating phenomena such as harmonic generation, sum- and difference-frequency generation, and self-phase modulation. We also developed formal properties of the nonlinear susceptibility. In this chapter, we develop in detail the consequences of second-order nonlinear polarization governed by $\chi^{(2)}$ for the generation of light at new frequencies, as well as the modification of the refractive index of a material by static voltages.

## 2.1 Wave equation description of nonlinear optics

Recall the fundamental idea that we described when taking about the anharmonic oscillator: a field with frequency components $\omega_1 \cdots \omega_n$ will lead to the creation of polarization at frequency $\omega_\sigma = \omega_1 + \cdots \omega_n$. Maxwell's equations tell us that polarization at frequency $\omega_\sigma$ generates a field at frequency $\omega_\sigma$. We will now make this explicit and concrete in this section by discussing Maxwell's equations in dielectric media. Then we will analyze a few canonical cases of second-order nonlinear interaction: sum-frequency generation, second-harmonic generation, difference-frequency generation, and the electro-optic effect. These effects form the modern backbone for *much* contemporary research in nonlinear optics.

In time-domain, Maxwell's equations can be combined to yield:

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, t) = -\mu_0 \partial_t (\mathbf{J}(\mathbf{r}, t) + \epsilon_0 \partial_t \mathbf{E}(\mathbf{r}, t)), \tag{2.1.1}$$

which, introducing the polarization via $\mathbf{J} = \partial_t \mathbf{P}$, can be written as

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, t) + \frac{1}{c^2} \partial_t^2 \mathbf{E}(\mathbf{r}, t) = -\mu_0 \partial_t^2 \mathbf{P}(\mathbf{r}, t). \tag{2.1.2}$$

Using the identity $\nabla \times \nabla \times = \nabla(\nabla \cdot) - \nabla^2$, we have

$$\nabla(\nabla \cdot \mathbf{E}(\mathbf{r}, t)) - \nabla^2 \mathbf{E}(\mathbf{r}, t) + \frac{1}{c^2} \partial_t^2 \mathbf{E}(\mathbf{r}, t) = -\mu_0 \partial_t^2 \mathbf{P}(\mathbf{r}, t). \tag{2.1.3}$$

Typically in linear electromagnetism you might be accustomed to dropping the $\nabla \cdot \mathbf{E}(\mathbf{r}, t)$ term because it vanishes: in nonlinear optics, it is nonzero due to the *anisotropy* of the

linear dielectric properties of the medium[1]. In fact, as we will learn in our discussion of sum-frequency generation, anisotropy is *required* to enforce a certain relation called phase-matching needed for *efficient* sum-frequency generation. *However*, the anisotropy of the linear dielectric properties is usually *small* - differences in the index of refraction in two directions can be less than 5% and so it leads to a small correction to the allowed polarizations of the electric field. Therefore, we will drop the divergence, leaving us with the equation:

$$\nabla^2\mathbf{E}(\mathbf{r},t) - \frac{1}{c^2}\partial_t^2\mathbf{E}(\mathbf{r},t) = \mu_0\partial_t^2\mathbf{P}(\mathbf{r},t). \tag{2.1.4}$$

In Fourier domain (in time), this may be written:

$$\nabla^2\mathbf{E}(\mathbf{r},\omega) - \frac{1}{c^2}\partial_t^2\mathbf{E}(\mathbf{r},\omega) = -\mu_0\omega^2\mathbf{P}(\mathbf{r},\omega). \tag{2.1.5}$$

Importantly, we see what we were saying: polarization at some frequency leads in general to a field at the same frequency.

It is customary to separate out the linear contribution to the polarization, writing the polarization as

$$\mathbf{P}(\mathbf{r},\omega) = \epsilon_0(\epsilon(\omega)-1)\mathbf{E}(\mathbf{r},\omega) + \mathbf{P}_{\mathrm{NL}}(\mathbf{r},\omega), \tag{2.1.6}$$

leaving us with the form of the equations we wish to use:

$$\nabla^2\mathbf{E}(\mathbf{r},\omega) + \epsilon(\omega)\frac{\omega^2}{c^2}\mathbf{E}(\mathbf{r},\omega) = -\mu_0\omega^2\mathbf{P}(\mathbf{r},\omega). \tag{2.1.7}$$

To proceed, we will need to analyze specific scenarios and geometries.

## 2.2 Sum-frequency generation

Consider a situation in which we have two plane waves of frequencies $\omega_1$ and $\omega_2$ impinge upon a second-order nonlinear crystal at normal incidence. They couple to this medium, and so at the input, there are two forward propagating waves $\mathbf{E}_1(\mathbf{r},t)$ and $\mathbf{E}_2(\mathbf{r},t)$. Taking their common direction of propagation as defining the $z$-direction, we may express the fields as

$$\mathbf{E}_i(\mathbf{r},t) = \hat{\epsilon}_i\left(A_i e^{ik_i z - i\omega_i t} + A_i^* e^{-ik_i z + i\omega_i t}\right), \tag{2.2.1}$$

where $A_i$ is the complex amplitude and $\hat{\epsilon}_i$ is the polarization. Now suppose that these two waves under sum-frequency generation, producing a third field $\mathbf{E}(\mathbf{r},t)$ in the $z$ direction with frequency $\omega_3 = \omega_1 + \omega_2$. The sum frequency wave *must* be in the $z$ direction. The reason is that the sum-frequency polarization is proportional to $e^{i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{r}}$ and so the wavevector of the generated field is $\mathbf{k}_1 + \mathbf{k}_2$. Assuming that fields 1 and 2 propagate in the $z$ direction, that forces the third to also propagate in the $z$ direction. That third-field can be expressed as

$$\mathbf{E}_3(\mathbf{r},t) = \hat{\epsilon}_3\left(A_3 e^{ik_3 z - i\omega_3 t} + A_3^* e^{-ik_3 z + i\omega_3 t}\right). \tag{2.2.2}$$

[1]In vacuum, this is because of Gauss' law, $\nabla\cdot\mathbf{D} = \nabla\cdot\epsilon\mathbf{E} = \rho_{\mathrm{free}}$ which tells us that the divergence of the displacement field vanishes in the absence of source charges. In an isotropic and uniform medium, $\nabla\cdot\mathbf{D} = \epsilon\nabla\cdot\mathbf{E}$ and so the electric field divergence vanishes. In an anisotropic medium, $\epsilon$ is different in at least two principal directions and so it cannot be factored out of the divergence.

As this sum-frequency field gets built-up from nonexistence, it must draw energy from the fields at frequency $\omega_1$ and $\omega_2$. We know that the intensity, which is the energy per unit time and area associated with the light field, goes as the square of the complex amplitude, and so the complex amplitudes of the two initial fields (1 and 2) must change.

To capture these changes, we make an ansatz of steady-state operation: the amplitudes are constant in time, but not in space. This describes a situation in which the field generated by sum-frequency generation propagates from the region in which it was generated. Since $z$ is the common direction of propagation, we write all three of our fields as

$$\mathbf{E}_i(\mathbf{r},t) = \hat{\epsilon}_i \left( A_i(z,t) e^{ik_i z - i\omega_i t} + A_i^*(z,t) e^{-ik_i z + i\omega_i t} \right), \tag{2.2.3}$$

and pursue an equation of motion for $A_i(z,t)$. In what follows, we will make a very common approximation in wave optics, which is the *slowly-varying envelope approximation* (SVEA): this approximation states that the normalized rate of the change in the amplitude envelope $|\partial_z A_i|/|A| \ll k_i |A|$. This is a reasonable approximation in most cases since the sum-frequency generating interaction is relatively inefficient and the amplitude changes at most by some tens of percent over often millimeters or centimeters of propagation. The SVEA manifests as follows: plug in our ansatz into the Maxwell equations: we then get

$$\left( \partial_z^2 A_i(z) e^{ik_i z} \hat{\epsilon}_i + \epsilon(\omega_i) \frac{\omega_i^2}{c^2} \hat{\epsilon}_i A_i(z,t) e^{ik_i z} \right) = -\mu_0 \omega_i^2 \mathbf{P}_i(z). \tag{2.2.4}$$

In writing this, we have (a) ignored transverse coordinate dependence of the fields (assuming plane waves in those directions), and (b) expressed our polarization in the form

$$\mathbf{P}_i(r,t) = \left( \mathbf{P}_i(z) e^{-i\omega_i t} + \mathbf{P}_i^*(z) e^{i\omega_i t} \right), \tag{2.2.5}$$

where I have made no assumption yet about the direction and wavevector of the polarization. Importantly, the second derivative can be expressed as

$$\partial_z^2 A_i(z) e^{ik_i z} = e^{ik_i z} \left( \partial_z^2 A + 2ik_i \partial_z A_i - k_i^2 A \right) \approx e^{ik_i z} \left( 2ik_i \partial_z A_i - k_i^2 A \right), \tag{2.2.6}$$

using the slowly-varying envelope approximation[2].

Putting it all together, we get (after some additional simplification)

$$2ik_i \partial_z A_{z,i} + \left( \hat{\epsilon}_i \cdot (\epsilon(\omega_i)\hat{\epsilon_i}) \frac{\omega_i^2}{c^2} - k_i^2 \right) A_i(z) = -\mu_0 \omega_i^2 \hat{\epsilon}_i \cdot \mathbf{P}(z) e^{-ik_i z}. \tag{2.2.7}$$

In what follows, we will neglect the anisotropy and pretend that the permittivity is a scalar such that $\epsilon(\omega_i)\omega_i^2/c^2 = k_i^2$ such that we are left with the equation[3].

$$\partial_z A_{z,i} = \frac{i\mu_0 \omega_i^2}{2k_i} \hat{\epsilon}_i \cdot \mathbf{P}(z) e^{-ik_i z}. \tag{2.2.8}$$

[2]Why didn't we also drop the first-derivative term?

[3]This is what is done in Boyd (he drops the tensorial nature of the permittivity): strictly speaking not very rigorous. One should have retained the divergence terms and then the non-derivative terms would have cancelled as the correct Maxwell equation for a plane wave polarized along some principle direction follows from the equation $-\mathbf{k} \times \mathbf{k} \times \hat{\epsilon} = \epsilon \hat{\epsilon} \omega_i^2/c^2$

We are now left with evaluating the nonlinear polarization. For the wave at frequency $\omega_3$ we may write

$$P_i(\omega_3) = \epsilon_0 \chi^{(2)}_{ijk}(\omega_3, \omega_1, \omega_2)\hat{\epsilon}_{3,i}\hat{\epsilon}_{1,j}\hat{\epsilon}_{2,k}A_1 A_2 e^{i(k_1+k_2)z}, \tag{2.2.9}$$

while for frequency $\omega_1$ we have

$$P_i(\omega_1) = \epsilon_0 \chi^{(2)}_{ijk}(\omega_1, \omega_3, -\omega_2)\hat{\epsilon}_{1,i}\hat{\epsilon}_{3,j}\hat{\epsilon}_{2,k}A_3 A_2^* e^{i(k_3-k_2)z}, \tag{2.2.10}$$

and for frequency $\omega_2$ we have

$$P_i(\omega_2) = \epsilon_0 \chi^{(2)}_{ijk}(\omega_2, \omega_3, -\omega_1)\hat{\epsilon}_{2,i}\hat{\epsilon}_{3,j}\hat{\epsilon}_{1,k}A_3 A_2^* e^{i(k_3-k_1)z}. \tag{2.2.11}$$

In what follows, we will consider the most important case of sum-frequency generation which is when the material is *lossless* (approximately) and thus dispersionless. Then, with full permutation symmetry, we find that

$$\hat{\epsilon}_i \cdot P_i(\omega_3) = 4\epsilon_0 d_{\text{eff}} A_1 A_2 e^{i(k_1+k_2)z}, \hat{\epsilon}_i \cdot P_i(\omega_{1,2}) = 4\epsilon_0 d_{\text{eff}} A_3 A^*_{2,1} e^{i(k_3-k_{2,1})z}. \tag{2.2.12}$$

Therefore, we may write

$$\partial_z A_3(z) = \frac{2i\omega_3^2 d_{\text{eff}}}{c^2 k_3} A_1 A_2 e^{i\Delta k z} \tag{2.2.13}$$

$$\partial_z A_1(z) = \frac{2i\omega_1^2 d_{\text{eff}}}{c^2 k_1} A_3 A_2^* e^{-i\Delta k z} \tag{2.2.14}$$

$$\partial_z A_2(z) = \frac{2i\omega_2^2 d_{\text{eff}}}{c^2 k_2} A_3 A_1^* e^{-i\Delta k z}, \tag{2.2.15}$$

where $\Delta k = k_1 + k_2 - k_3$. These are the equations of sum-frequency generation.

### 2.2.1 Sum-frequency generation in the non-depleted approximation

In general these equations are nonlinear and difficult to solve (there is an exact solution). That said, most of the insight comes from the regime when the nonlinearity is weak and the intensity built up in the sum-frequency is much smaller than the intensity in the driving fields. Then, we may approximate the driving fields as constant in space. This is the non-depleted approximation. Then, we may integrate the equation for $A_3$ directly up to a length $z = L$, finding

$$A_3(L) = \frac{2i\omega_3^2 d_{\text{eff}}}{c^2 k_3} A_1(0) A_2(0) \frac{e^{i\Delta k L} - 1}{i\Delta k} = \frac{2i\omega_3^2 d_{\text{eff}}}{c^2 k_3} A_1(0) A_2(0) e^{i\Delta k L/2} L \text{sinc}\left(\frac{1}{2}\Delta k L\right). \tag{2.2.16}$$

The intensity in the sum-frequency wave is then given from Poynting's theorem as $2\epsilon_0 n_3 c|A_3|^2$, in terms of the intensities of the incident waves, as

$$I_3(L) = \frac{2\omega_3^2 d_{\text{eff}}}{n_1 n_2 n_3 \epsilon_0 c^3} I_1(0) I_2(0) L^2 \text{sinc}^2\left(\frac{1}{2}\Delta k L\right). \tag{2.2.17}$$

Importantly, the intensity in the sum-frequency field is proportional to the product of the intensities of the incident fields, quadratic in propagation length, and weighted by the sinc factor, which rapidly decays if $\Delta kL \gg 1$. The sinc factor is maximized when $\Delta k = 0$ or equivalently when

$$k_1 + k_2 = k_3. \tag{2.2.18}$$

Of course, by construction, it is also the case that

$$\omega_1 + \omega_2 = \omega_3. \tag{2.2.19}$$

This condition is called the *phase matching* condition and in a photon picture could be thought of as an expression of energy-momentum conservation. It is clear that to have efficient generation of the sum frequency, we must achieve phase-matching. While we have only derived this in the perturbative limit, it holds generally.

### 2.2.2 Phase-matching

The phase-matching condition is not automatically satisfied and in fact takes some doing to satisfy, and it can be satisfied using different techniques. We discuss this here. The wavevector condition can be expressed as

$$n(\omega_1)\omega_1 + n(\omega_2)\omega_2 = n(\omega_3)\omega_3 = n(\omega_3)(\omega_1 + \omega_2). \tag{2.2.20}$$

Combining this with the frequency-matching condition, we have

$$\frac{n(\omega_1)\omega_1 + n(\omega_2)\omega_2}{\omega_1 + \omega_2} = n(\omega_3). \tag{2.2.21}$$

This equation is in general *not satisfied*. For example, many materials present *positive dispersion* meaning that the index increases with frequency. In the infrared and visible range, this is typical. Let us assume that the material is isotropic and is thus described only by one index of refraction at each frequency. In a system as described, phase-matching is impossible to satisfy, precluding efficient nonlinearities. Suppose without loss of generality that $\omega_3 > \omega_2 > \omega_1$. Then for positive dispersion, we also have $n(\omega_3) > n(\omega_2) > n(\omega_1)$. The expression on the left-hand side of the above equation is the frequency-weighted average index of refraction between waves 1 and 2. There is no way for this quantity to be larger than $n(\omega_2)$, and therefore cannot be equal to $n(\omega_3)$.

### 2.2.3 Birefringent phase matching: angle and temperature tuning

One way to deal with phase-matching is to use *anomalous dispersion*: near a resonance of a material, the index can decrease with increasing frequency. This is not the most common solution that is used: instead, we typically exploit the natural *anisotropy* of a system, the fact that the index of refraction is different in different directions. Let us analyze the example that Boyd does, of a material in the $3m$ (trigonal) crystal class. For a trigonal system, we have that there are two distinct principal values of the index of refraction, $n_e$ ($e$ for extraordinary) corresponding to field polarization along the crystal or $c$ axis of the system, and $n_o$ in the two directions perpendicular to the $c$-axis. We call such a system *uniaxial* because there is one

principal axis with a distinct index of refraction (the c-axis), while the other two are equivalent. A biaxial system refers to the case where all three principal directions have different indices of refraction.

For a wave propagating along wavevector $\mathbf{k}$ at an angle $\theta$ to the $c$-axis, the effective index seen (as shown on Problem Set 1) is

$$\frac{1}{n^2(\theta)} = \frac{\sin^2\theta}{n_e^2} + \frac{\cos^2\theta}{n_o^2}. \tag{2.2.22}$$

Importantly, the index of refraction associated with extraordinary polarization and ordinary polarization can be different. Ordinary polarization refers to the field polarization (technically D-field) being orthogonal to the c-axis, while extraordinary polarization refers to the other polarization state which has a nonzero projection with the c-axis. In a *negative* uniaxial system, $n_e < n_o$ while in a *positive* uniaxial system $n_e > n_o$.

How does this help with phase matching? If all three waves in the SFG process have ordinary polarization, we can't phase-match, assuming positive dispersion. If all three waves in the SFG process have extraordinary polarization, we also cannot phase-match in the case where all three waves propagate in the same direction (collinear SFG). It is the same argument as in the isotropic case.

However, we see from the argument we made in the isotropic case that if we could somehow make $n_3$ lower than $n_2$, phase-matching could at least be possible. This is the idea behind *type-I phase-matching*, which refers to the case where the two input waves have ordinary polarization and the sum frequency wave has extraordinary polarization (in a negative uniaxial crystal[4]). You might also imagine in the case of the negative uniaxial crystal that I could get away with making one of the two input polarizations extraordinary (the lower-frequency input would be easier because it pulls the weighted average down less). That is the case of *type-II phase matching*[5].

Let us now see how this all shakes out mathematically. Let us consider type-I phase matching for second-harmonic generation ($\omega + \omega \to 2\omega$). You can convince yourself that the phase matching condition is simply (in the negative uniaxial case)

$$2\omega n_o(\omega) \to 2\omega n_e(2\omega) \implies n_o(\omega) = n(2\omega, \theta). \tag{2.2.23}$$

If we take our three waves to have the same direction, which is at an angle $\theta$ to the $c$-axis, then our phase matching condition becomes

$$\frac{1}{n_0^2(\omega)} = \frac{\sin^2\theta}{n_e^2(2\omega)} + \frac{\cos^2\theta}{n_o^2(2\omega)}. \tag{2.2.24}$$

We may express this as

$$\sin^2\theta = \frac{\frac{1}{n_o^2(\omega)} - \frac{1}{n_o^2(2\omega)}}{\frac{1}{n_e^2(2\omega)} - \frac{1}{n_o^2(2\omega)}}. \tag{2.2.25}$$

Let's sanity check this. Phase-matching implies that this equation can be solved for some angle, meaning that the right hand side is between 0 and 1. If the right-hand side becomes

[4] In a positive uniaxial crystal, we reverse the polarizations (inputs are extraordinary, output is ordinary

[5] Again, for positive uniaxial crystals, reverse the polarizations

negative, or larger than 1, we can't phase-match. We know that lots of normal dispersion makes phase-matching harder, and we know that birefringence makes things easier. Note that by assumption that we have normal dispersion, the numerator is positive. Similarly, assuming a negative uniaxial crystal implies that the denominator is positive. If the normal dispersion is strong, then the numerator increases, pushing us away from having a solution. Similarly, if the birefringence is weak, then the denominator gets small, having the same effect of pushing us away from having a solution. So we see that we have a kind of ratio of dispersion to birefringence.

Of course, for a given frequency, the principal indices of refraction as a function of frequency is fixed, and so we can think of the right-hand side as fixed. By rotating the crystal however, we change the angle of the $c$-axis to our incident light direction, and so we can think of performing *angle tuning* until phase-matching is satisfied.

There is a severe drawback related to angle tuning, which is spatial walk-off (in Problem Set 2, you'll analyze temporal walk off of pulses). In reality, we don't send plane waves at a crystal, we send localized wavepackets. You might imagine then that if the two input field wavepackets and the sum-frequency wavepacket do not overlap in space, there should not be a nonlinear interaction since the nonlinear polarization cares about the three fields at the same location. Let us consider again the case of type-I phase-matching for second-harmonic generation. The fundamental (input) wave has a group velocity along the direction of $\mathbf{k}$. The harmonic however, does not[6]. As a result, the harmonic moves in a different direction and speed from the fundamental and so after some "drift time" they will not overlap, ceasing the nonlinear interaction. This is called *walk-off* for reasons that are sufficiently self-explanatory.

This can be avoided by *temperature tuning*. By changing the temperature of the medium, we can change $n_{o,e}(\omega)$ and $n_{o,e}(2\omega)$ to satisfy the phase-matching equation for angles $\theta$ for which walk-off does not occur. Those angles are, as per the previous footnote, $\theta = 0$ and $\theta = \pi/2$. Of course, $\theta = 0$ cannot get you phase-matching by assumption that there is normal dispersion (even with temperature tuning). Importantly, for temperature tuning to work, we need the birefringence to respond strongly to temperature while the dispersion responds less. The opposite case would suppress phase-matching.

### 2.2.4 Quasi-phase matching

There is another powerful trick for achieving phase matching, involving in principle no temperature-tuning, no birefringence, and using collinear fields. This is of course very helpful as it gets around a bunch of the issues mentioned above! It also allows access to elements of the $d$ tensor that are often the largest. For example, in some materials, including lithium niobate, $d_{33}$ is the largest element. However, if the polarizations of the three waves in SFG or the two polarizations in SHG are not the same, then $d_{33}$ does not contribute to $d_{\text{eff}}$. Of course, if the polarizations are the same, we run into issues related to normal dispersion.

The solution is to periodically flip the sign of $d_{\text{eff}}$ (equivalently, flipping the sign of $\chi^{(2)}$. How do we do this? From the standpoint of the cubic oscillator we analyzed, the sign of $\chi^{(2)}$ is controlled by $a$ the constant setting the cubic contribution to the potential felt by the

[6] In an anisotropic medium, the group velocity is $\nabla_{\mathbf{k}}\omega(\mathbf{k})$. For the extraordinary polarization, we can take the result in Problem 1 and write $2\omega_{\mathbf{k}}\nabla_{\mathbf{k}}\omega_{\mathbf{k}} = c^2\left(\hat{k}2k\left(\frac{\sin^2\theta}{n_e^2}+\frac{\cos^2\theta}{n_o^2}\right)+\hat{\theta}k\frac{\sin 2\theta}{n_e^2}-\frac{\sin 2\theta}{n_o^2}\right)$. This clearly has a nonzero component along $\hat{k}$ unless $\theta = 0$ or $\theta = \pi/2$

oscillator. It turns out that in centrosymmetry breaking materials like lithium niobate, they are *ferroelectric* having a permanent electric dipole moment not unlike the permanent magnetic moment of a ferromagnet like iron. You can imagine that the presence of frozen permanent dipoles in a material creates a strong potential for electrons, and is solely responsible for breaking centrosymmetry[7]! The direction of these dipoles can be reversed by application of an electric field, thus flipping the sign of the symmetry-breaking potential and this is effectively "flipping $a$". So our plan is to periodically flip the polarity (in a way to be elaborated) of our ferroelectric: this is called *periodic poling*[8].

Physically, why does this help? Suppose we have a segment of nonlinear material of fixed polarity, and assume we're doing SFG where all three polarizations are the same in a normally dispersive material. As we have established already, we cannot phase match. Therefore, $\Delta k \neq 0$, and we expect to see that, in the case of weak depletion, $A_3$ is given by (2.2.16) and will first rise but then come back down. Suppose that when $|A_3|$ reaches its maximum at $z = \pi/\Delta k$, we suddenly flip the sign of $d$: then $A_3$ will increase instead of decrease. It will basically "copy" its evolution from 0 to $z$. When that branch reaches its maximum, flip the sign of $d$ again. This tells us automatically what the period of our poling variation should be. If the period of $\Lambda$ is

$$\Lambda = \frac{2\pi}{\Delta k}, \tag{2.2.26}$$

then we're in business. Our graphical argument however suggests that we could let $\frac{1}{2}\Delta k z = (2m+1)\pi/2$ before flipping the poling direction, allowing also for periods $(4m+2)\pi/\Delta k$.

Let us suppose now that we have a periodic $d_{\text{eff}}$ in the equations for SFG that we have developed (with period $\Lambda$). Then it may be written as

$$d_{\text{eff}}(z) = \sum_{m=-\infty}^{\infty} d_m e^{2\pi i m/\Lambda}. \tag{2.2.27}$$

In what follows, suppose one particular harmonic $q$ allows for phase-matching. We take only that harmonic[9], thus approximating $d_{\text{eff}}(z) \approx d_q e^{2\pi i q/\Lambda} + d_q^* e^{-2\pi i q/\Lambda}$. The coupled amplitude equations become:

$$\partial_z A_3(z) = \frac{2i\omega_3^2 d_q}{c^2 k_3} A_1 A_2 e^{i\Delta k_q z} \tag{2.2.28}$$

$$\partial_z A_1(z) = \frac{2i\omega_1^2 d_q^*}{c^2 k_1} A_3 A_2^* e^{-i\Delta k_q z} \tag{2.2.29}$$

$$\partial_z A_2(z) = \frac{2i\omega_2^2 d_q^*}{c^2 k_2} A_3 A_1^* e^{-i\Delta k_q z}, \tag{2.2.30}$$

where $\Delta k_m = k_1 + k_2 - k_3 + 2\pi q/\Lambda$. Clearly, in the non-depleted pump case, $A_3$ has a chance to build up coherently over long distances if one of these $\Delta k_m$ terms is zero.

[7] At high temperatures, LN is a cubic paraelectric and is thus centrosymmetric and doesn't have second-order nonlinearity. This also tells us that we can probe symmetry-breaking phases of matter by their second-harmonic / nonlinear response.

[8] Terms in nonlinear optics make a lot of sense about 98% of the time.

[9] The other terms give a small oscillatory contribution that could be ignored.

### 2.2.5 Manley-Rowe relations

Let us go back to our analysis of the coupled-wave equations for sum-frequency generation. We were able to simply solve these equations in the undepleted approximation. But let us consider what physical statements are true outside of this approximation.

Clearly, we expect energy conservation to hold, which indicates that in a steady-state situation, the total intensity of all three waves is invariant under propagation if no energy builds up in any sub-region of the system. Therefore, we expect that

$$\frac{d}{dz}(I_1 + I_2 + I_3) = 0. \tag{2.2.31}$$

For continuous-wave light, the time-averaged intensity of any one wave is $I_i = 2\epsilon_0 n_i c|A_i|^2$[10] Therefore, we expect to see that

$$\frac{d}{dz}\sum_i n_i|A_i|^2 = 2\sum_i n_i \text{Re } A_i^* \frac{dA_i}{dz} = 0. \tag{2.2.32}$$

This may easily be checked to be true of the coupled-amplitude equations. In particular, when calculating the $z$-derivative of the intensities, you would find that

$$\frac{dI_1}{dz} = 2\epsilon_0 c \frac{2\omega_1 d_{\text{eff}}}{c} \text{Re } iA_1^* A_2^* A_3 e^{-i\Delta k z}, \tag{2.2.33}$$

$$\frac{dI_2}{dz} = 2\epsilon_0 c \frac{2\omega_2 d_{\text{eff}}}{c} \text{Re } iA_1^* A_2^* A_3 e^{-i\Delta k z}, \tag{2.2.34}$$

and

$$\frac{dI_3}{dz} = -2\epsilon_0 c \frac{2\omega_3 d_{\text{eff}}}{c} \text{Re } iA_1 A_2 A_3^* e^{i\Delta k z}. \tag{2.2.35}$$

The sum of these terms vanishes if $\omega_1 + \omega_2 - \omega_3 = 0$, which is true by construction. In writing this, we notice another set of exact relations, namely:

$$\frac{dM_{1,2}}{dz} \equiv \frac{1}{\omega_1}\frac{dI_1}{dz} - \frac{1}{\omega_2}\frac{dI_2}{dz} = 0, \tag{2.2.36}$$

$$\frac{dM_{1,3}}{dz} \equiv \frac{1}{\omega_1}\frac{dI_1}{dz} + \frac{1}{\omega_3}\frac{dI_3}{dz} = 0, \tag{2.2.37}$$

and

$$\frac{dM_{2,3}}{dz} \equiv \frac{1}{\omega_2}\frac{dI_2}{dz} + \frac{1}{\omega_3}\frac{dI_3}{dz} = 0. \tag{2.2.38}$$

These relations are called *Manley-Rowe* relations, and indicate that the following quantities are invariant:

$$M_{1,2} = \frac{I_1}{\omega_1} - \frac{I_2}{\omega_2}, M_{1,3} = \frac{I_1}{\omega_1} + \frac{I_3}{\omega_3}, M_{2,3} = \frac{I_2}{\omega_2} + \frac{I_3}{\omega_3}. \tag{2.2.39}$$

Invariants are physically useful quantities and often correspond to quantities with clear physical interpretation such as energy, momentum, angular momentum, etc. The Manley-Rowe invariants correspond to conservation of photon number. In particular, let us consider

[10] This is assuming that we specify a CW field as $E_i = A_i e^{-i\omega_i t} + A_i^* e^{i\omega_i t}$.

the photon picture of sum frequency generation. In sum frequency generation, we have one photon at $\omega_1$ is annihilated, one photon at $\omega_2$ is annihilated, and one photon at $\omega_3$ is created. Let us imagine that as time elapses, sum-frequency generation processes occur. Then we certainly expect some type of deterministic relation between changes in the number of one type of photons and photons of another. If the number of photons per unit length at frequencies $\omega_{1,2,3}$ is $n_{1,2,3}$ respectively, then we expect that

$$\frac{dn_1}{dt} = \frac{dn_2}{dt} = -\frac{dn_3}{dt}, \tag{2.2.40}$$

where the number of photons per unit volume at frequencies $\omega_{1,2,3}$ is $n_{1,2,3}$. To relate this to $z$ derivatives, remember that these photons are all moving with their own velocities $v_i$ (the speed of light divided by the index of refraction). Therefore the time derivative is in fact $d/dt = \partial/\partial_t + v_i\partial/\partial_z$. In the steady-state assumption, the partial time derivative vanishes and we are left with $v_1\partial_z n_1 = v_2\partial_z n_2$. But the intensity is simply $\hbar\omega_i v_i n_i$, and so we see simply that this is equivalent to $\partial_z M_{1,2} = 0$, which is the same as above[11]

### 2.2.6 Sum-frequency generation beyond the non-depleted approximation

Let us consider a situation of sum-frequency generation in which the lowest frequency wave at frequency $\omega_1$ is weak and the second input wave at frequency $\omega_2$ is strong. We could imagine the $\omega_1$ field to be an infrared field and the $\omega_2$ field to be a laser field, and they mix to create a wave at frequency $\omega_3 = \omega_1 + \omega_2$. It is possible to solve the equations of sum-frequency generation exactly, in terms of Jacobi elliptic functions. However, when the $\omega_2$ field is strong, we can treat it as undepleted, while still tracking the spatial evolution of the $\omega_1$ and $\omega_3$ fields. For the fields at $\omega_1$ and $\omega_3$, we have

$$\partial_z A_3(z) = \frac{2i\omega_3^2 d_{\text{eff}}}{c^2 k_3} A_1 A_2 e^{i\Delta k z} \tag{2.2.41}$$

$$\partial_z A_1(z) = \frac{2i\omega_1^2 d_{\text{eff}}}{c^2 k_1} A_3 A_2^* e^{-i\Delta k z} \tag{2.2.42}$$

When $A_2$ is treated as constant, we see that we have two linear differential equations with spatially varying coefficients which can be solved. Before doing so, it will help to define $A_3(z) = \tilde{A}_3(z)e^{i\Delta k z/2}$ and $A_1(z) = \tilde{A}_1(z)e^{-i\Delta k z/2}$. It immediately follows that

$$\partial_z \tilde{A}_3(z) + \frac{i\Delta k}{2}\tilde{A}_3(z) = \alpha \tilde{A}_1 \tag{2.2.43}$$

and

$$\partial_z \tilde{A}_1(z) - \frac{i\Delta k}{2}\tilde{A}_1(z) = \beta \tilde{A}_3 \tag{2.2.44}$$

with $\alpha = \frac{2i\omega_3^2 d_{\text{eff}}}{c^2 k_3} A_2$ and $\beta = \frac{2i\omega_1^2 d_{\text{eff}}}{c^2 k_1} A_2^*$. This may be written in matrix notation as

$$\partial_z \begin{pmatrix} \tilde{A}_1 \\ \tilde{A}_3 \end{pmatrix} = \begin{pmatrix} \frac{1}{2}i\Delta k & \beta \\ \alpha & -\frac{1}{2}i\Delta k \end{pmatrix} \begin{pmatrix} \tilde{A}_1 \\ \tilde{A}_3 \end{pmatrix}. \tag{2.2.45}$$

[11] Here I have a partial derivative, that is because I explicitly noted the time dynamics of the photons moving. In the above, there was no notion of time as we took the steady-state limit of the wave equation: thus the total and partial derivatives are interchangeable.

We know from systems of differential equations such as these that the eigenvalues of the matrix on the right hand side dictate the $z$-evolution of the fields. The two eigenvalues denoted $\lambda_{1,2}$ can be found to be

$$\lambda_{1,2} = \pm i\sqrt{\frac{4\omega_1^2\omega_3^2 d_{\text{eff}}^2}{k_1 k_3 c^4}|A_2|^2 + \frac{(\Delta k)^2}{4}} \equiv \pm\lambda, \tag{2.2.46}$$

with $\lambda_1, \lambda_2$ being assigned the positive (negative) roots respectively. We may then write

$$\tilde{A}_{1,3}(z) = J_{1,3}\cos(\lambda z) + K_{1,3}\sin(\lambda z), \tag{2.2.47}$$

with $J, K$ being constants.

Let us take as initial conditions the typical case in which there is no initial sum-frequency field, and so we have $J_1 = A_1(0)$ and $K_1 = \frac{i\Delta k}{2}A_1(0)$, while $J_3 = 0$ and $K_3 = \frac{2i\omega_3^2 d_{\text{eff}}}{c^2 k_3}A_2 A_1(0)$. Putting it all together, we have

$$A_1(z) = \left[\cos(\lambda z) + \frac{i\Delta k}{2\lambda}\sin(\lambda z)\right]A_1(0)e^{-i\Delta k z/2} \tag{2.2.48}$$

and

$$A_3(z) = \left[\frac{2i\omega_3^2 d_{\text{eff}}}{c^2 k_3 \lambda}A_2\sin(\lambda z)\right]A_1(0)e^{i\Delta k z/2}. \tag{2.2.49}$$

As can be seen from these expressions, the sum-frequency wave builds up from zero, oscillating in space at period $2\pi/\lambda$. The maximum intensity of the sum-frequency field is

$$I_{3,\max} = \frac{n_3}{n_1}\frac{\frac{4\omega_3^2 d_{\text{eff}}^2|A_2|^2}{c^4 k_3^2}}{\frac{4\omega_1^2\omega_3^2 d_{\text{eff}}^2}{k_1 k_3 c^4}|A_2|^2 + \frac{(\Delta k)^2}{4}}I_1(0). \tag{2.2.50}$$

It is clear from this expression that the maximum is realized when phase-matching is satisfied ($\Delta k = 0$), and we have

$$I_{3,\max} = \frac{\omega_3}{\omega_1}I_1(0). \tag{2.2.51}$$

## 2.3 Second-harmonic generation

In second-harmonic generation, we take a wave at frequency $\omega_1$ and convert it using a second-order nonlinear medium into a wave at frequency $\omega_2 = 2\omega_1$. In the photon picture, we take two photons at $\omega_1$ and convert it into a photon at $\omega_2$. The physics of second-harmonic generation is similar to that of sum-frequency generation: indeed we can see second-harmonic generation as a limiting case of sum-frequency generation where the two input frequencies are equal. The coupled-amplitude equations can be written as (Reader: make sure to derive this!)

$$\partial_z A_1(z) = \frac{2i\omega_1^2 d_{\text{eff}}}{c^2 k_1}A_1^* A_2 e^{-i\Delta k z} \tag{2.3.1}$$

$$\partial_z A_2(z) = \frac{i\omega_2^2 d_{\text{eff}}}{c^2 k_2}A_1^2 e^{i\Delta k z}, \tag{2.3.2}$$

with $\Delta k = 2k_1 - k_2$[12] It is simple to follow the discussion of sum-frequency generation and find the second-harmonic intensity in the undepleted pump approximation for the fundamental, and the reader is very strongly urged to do this. Here, we will solve this equation without the non-depleted pump approximation.

In what follows, we will consider the simpler case of these equations where the interaction is phase-matched: this gives the most efficient second-harmonic conversion anyway. Therefore we may set $\Delta k = 0$. To solve this equation, we start by identifying useful invariants. One such invariant is the total intensity, which implies that

$$n_1|A_1|^2 + n_2|A_2|^2 = \text{const.}. \tag{2.3.3}$$

The Manley-Rowe invariant for second-harmonic generation is identical to the total intensity (reader: check that!). There is one other quantity which is invariant when phase-matching holds. It is

$$\text{Re } A_1^2 A_2^* = |A_1|^2|A_2|\cos(2\phi_1 - \phi_2) = \text{const.}, \tag{2.3.4}$$

where we have defined $A_1 = |A_1|e^{i\phi_1}$ and $A_2 = |A_2|e^{i\phi_2}$. This invariant may seem mysterious at first glance (and indeed Boyd just finds this from the coupled amplitude equations directly), but corresponds to constancy of the time-averaged energy density[13].

We will consider the simple but very common case where there is no initial intensity in the second harmonic wave (other cases can easily be done but it involves more steps). In this case, it is clear that the phase of the incident wave does not matter (absolute phases can always be multiplied away!): we set $\phi_1(0)$ to zero without loss of generality. Our nonlinear invariant tells us that $|A_1|^2|A_2|\cos(2\phi_1 - \phi_2) = 0$ which means that $\cos(2\phi_1 - \phi_2) = 0$ unless we are at locations $z$ for which $|A_2| = 0$ in which case the phase $\phi_2$ loses any meaning anyway and $\phi_1$ can be set arbitrarily. Therefore, we see that

$$2\phi_1 - \phi_2 = (2m+1)\frac{\pi}{2} \tag{2.3.5}$$

for any integer $m$. If convert the evolution equation for $A_2$ into equations for $|A_2|$ and $\phi_2$, we find that

$$\partial_z|A_2| + i|A_2|\partial_z\phi_2 = \frac{i\omega_2^2 d_{\text{eff}}}{k_2 c^2}|A_1|^2 e^{i(2\phi_1 - \phi_2)}, \tag{2.3.6}$$

[12]Note that the "missing" factor of 2 in the equation for the second harmonic field comes from the fact that there is only one instance of $A_1^2$ upon squaring the electric field, as opposed to the two instances of $A_1^* A_2$.

[13]The linear part of the time-averaged energy density is simply the time-average of $\sum_i \epsilon_0 n_i^2 E_i^2$ which is $2\epsilon_0\left(n_1^2|A_1|^2 + n_2^2|A_2|^2\right)$. The nonlinear energy density is proportional to $\chi^{(2)}E^3$ as per our discussion of lossless media in chapter 1. The time-averaged part corresponds to the DC terms in $E^3 = (A_1 e^{ik_1 z - i\omega_1 t} + A_2 e^{ik_2 z - i\omega_2 t} + \text{c.c.})^3$. The frequencies in this expression are $\pm\omega_1 \pm \omega_1 \pm \omega_2 = \pm\omega_1 \pm \omega_1 \pm 2\omega_1$. Clearly there are only two terms for which the sum of these is zero. The time-averaged nonlinear energy density associated with those terms is $A_1^2 A_2^* e^{i\Delta kz} + A_1^{*2} A_2 e^{-i\Delta kz} = 2|A_1|^2|A_2|\cos(2\phi_1 - \phi_2 + \Delta kz)$. The total time-averaged energy density is the sum of these two terms. In the steady-state, constancy of the time-averaged energy density $u$: $du/dt = 0$ implies that $\partial_z u = 0$. At the same time, we also know that the time-averaged *intensity* is conserved as a function of $z$: $n_1|A_1|^2 + n_2|A_2|^2 = \text{const.}$. If phase-matching holds, then $n_1 = n_2$ implying $|A_1|^2 + |A_2|^2$ is conserved and so similarly, the linear part of the time-averaged energy density is conserved. Thus, for the time-averaged energy density to be constant, we require $|A_1|^2|A_2|\cos(2\phi_1 - \phi_2)$ to be a constant, as claimed. Note that if phase-matching is not satisfied, then constancy of the intensity does not imply constancy of the linear part of the energy density, and so things get more complicated. It turns out that these invariants are *much* easier to get at quantum mechanically as there it just corresponds to the fact that the expectation value of energy in a time-independent Hamiltonian is conserved!

implying that

$$\partial_z |A_2| = -\frac{\omega_2^2 d_{\text{eff}}}{k_2 c^2} |A_1|^2 \sin(2\phi_1 - \phi_2). \tag{2.3.7}$$

This may be expressed in terms of $|A_2|$ alone via

$$\partial_z |A_2| = -\frac{\omega_2^2 d_{\text{eff}}}{k_2 c^2} (C - |A_2|^2) \sin(2\phi_1 - \phi_2), \tag{2.3.8}$$

where under phase-matching we have $|A_1|^2 + |A_2|^2 = C = |A_1(0)|^2$. Note that the nonlinear invariant vanishing tells us that the sine is either 1 or -1. It is clear that since at first, the intensity in $I_2$ must increase (intensity is nonnegative), the sine must be -1. We then find that

$$\int_0^{|A_2(z)|} \frac{dx}{|A_1(0)|^2 - x^2} = \frac{2\omega_1 d_{\text{eff}}}{n_1 c} z, \tag{2.3.9}$$

which using standard logarithmic integral identities can be reduced to

$$|A_2(z)| = |A_1(0)| \tanh\left(\frac{\omega_1 d_{\text{eff}} |A_1(0)|}{n_1 c} z\right). \tag{2.3.10}$$

In this case, the second-harmonic field is capable of capturing all of the incident intensity, corresponding to complete conversion of the fundamental wave. This behavior is in some sense non-representative of the full solution space: if the nonlinear invariant is finite, or if phase-matching is not satisfied, one will have an oscillatory power exchange between the fundamental and the second harmonic.

## 2.4 Difference-frequency generation and parametric generation

Maxwell's equations in a lossless medium (even in the nonlinear case) are reversible: playing the solutions backwards generates another valid solution[14]. That suggests that if we take a phenomenon like sum-frequency generation, and play it in reverse, we would see an effect in which a field at frequency $\omega_3$ generates fields at frequencies $\omega_1, \omega_2$ ($\omega_1 < \omega_2 < \omega_3$): this is called difference-frequency generation (or non-degenerate parametric down-conversion, depending on initial conditions). Similarly, if we watch second-harmonic generation in reverse, we would see a field at frequency $2\omega_1$ generate fields at frequency $\omega_1$, an effect called degenerate parametric down-conversion. Let us study the case of non-degenerate parametric down-conversion.

The relevant coupled amplitude equations are *exactly* those describing sum-frequency generation. Let us suppose however that there is a strong incident wave at frequency $\omega_3$ which is treated as un-depleted. The fields $A_1$ and $A_2$ are "weak" and their spatial variations are taken to be non-negligible. Then, the resulting linear equations for $A_1, A_2$ are:

$$\partial_z A_1(z) = \frac{2i\omega_1^2 d_{\text{eff}}}{c^2 k_1} A_3 A_2^* e^{-i\Delta k z} \tag{2.4.1}$$

[14]Why lossless? Consider an absorbing medium which extracts energy irreversibly from a wave in time. If we play the dynamics backwards, the wave gets amplified, which only occurs if the medium is active. If we watch a movie of the time-reversed solution but know that all of our materials are passive, then the resulting solution is invalid.

$$\partial_z A_2(z) = \frac{2i\omega_2^2 d_{\text{eff}}}{c^2 k_2} A_3 A_1^* e^{-i\Delta k z}, \tag{2.4.2}$$

where $\Delta k = k_1 + k_2 - k_3$. These equations are *almost* directly solvable linear equations. The "issue" is that the fields couple to their conjugates, which are independent variables[15]. This can of course be quickly fixed by conjugating the second equation, yielding:

$$\partial_z A_1(z) = \frac{2i\omega_1^2 d_{\text{eff}}}{c^2 k_1} A_3 A_2^* e^{-i\Delta k z} \tag{2.4.3}$$

$$\partial_z A_2^*(z) = -\frac{2i\omega_2^2 d_{\text{eff}}}{c^2 k_2} A_3^* A_1 e^{i\Delta k z}. \tag{2.4.4}$$

We can solve this in exactly the same way that we solved sum-frequency generation with one strong undepleted wave. We define $A_1 = \tilde{A}_1 e^{-i\Delta k z/2}$ and $A_2^* = \tilde{A}_2^* e^{i\Delta k z/2}$ and convert into a matrix differential equation with constant coefficients. It immediately follows that

$$\tilde{A}_1(z) = J_1 \cosh(\lambda z) + K_1 \sinh(\lambda z), \tag{2.4.5}$$

while

$$\tilde{A}_2^*(z) = J_2 \cosh(\lambda z) + K_2 \sinh(\lambda z), \tag{2.4.6}$$

with

$$\lambda = \pm\sqrt{\frac{4\omega_1^2\omega_2^2 d_{\text{eff}}^2}{k_1 k_2 c^4}|A_3|^2 - \frac{(\Delta k)^2}{4}}. \tag{2.4.7}$$

If we take for example that $A_2(0) = 0$, then matching boundary conditions would give

$$A_1(z) = \left[\cosh(\lambda z) + \frac{i\Delta k}{2\lambda}\sinh(\lambda z)\right] A_1(0) e^{-i\Delta k z/2}, \tag{2.4.8}$$

and

$$A_2^*(z) = \left[-\frac{2i\omega_2^2 d_{\text{eff}} A_3^*(0)}{c^2 k_2 \lambda}\sinh(\lambda z)\right] A_1(0) e^{i\Delta k z/2}. \tag{2.4.9}$$

In the phase-matched case, this reduces to

$$A_1(z) = A_1(0)\cosh(\lambda z), \tag{2.4.10}$$

and

$$A_2(z) = i\sqrt{\frac{n_1\omega_2}{n_2\omega_1}} e^{i\phi_3} A_1^*(0)\sinh(\lambda z), \tag{2.4.11}$$

where $\phi_3 = A_3/|A_3|$. For distances $z \gg \lambda$, we may easily see that the intensities of both lower-frequency waves grows exponentially as $e^{2\lambda z}$, representing a type of exponential gain not unlike that of a laser amplifier. Unlike the laser amplifier however, the energy of the amplified wave is not extracted from excited electrons in atoms or semiconductors, but instead by extracting energy from another light wave coherently through nonlinear polarization[16].

[15] This is very important. We know that for complex variables, we can treat the real and imaginary parts of the variable as independent. Therefore, we can also treat a complex variable and its conjugate as independent *single variables* since they are related to the real and imaginary part by a linear transformation!

[16] One might wonder how: if this process is related by inverse to sum-frequency generation, why do we have exponential amplification rather than oscillation? We leave this question for the reader to think about!

## 2.5 Parametric oscillation

The fact that we can amplify waves using parametric interactions forms the basis for a power type of laser-like light source called the optical parametric oscillator (OPO). Such parametric oscillators have become extremely important alternatives to "conventional" lasers based on excited media due to their very widely tunable wavelengths, and have accordingly made an important commercial impact. In this section, we will analyze some of the basic behaviors of the parametric oscillator.

Let us consider a resonator formed by two mirrors of complex amplitude reflectivities $r_1$ and $r_2$, defined such that the transmitted energy fraction is $T_i = 1 - R_i \equiv 1 - |r_i|^2$. Inside this resonator is a second-order nonlinear medium. We assume that the mirrors are perfectly transmissive for the pump field at frequency $\omega_3$. If the mirrors are only highly reflective for one of the lower-frequency waves, we call that wave the signal, and the OPO is a singly-resonant OPO. If the mirrors are highly reflective for both lower-frequency waves, the OPO is doubly-resonant and the name signal is used for the frequency we are more interested in, and idler for the frequency we are less interested in.

If the mirror reflectivities are high for the signal, then the signal can bounce around in this resonator (also called a cavity) many times, effectively experiencing an enhanced length of nonlinear interaction and enabling more efficient extraction of energy from the pump. At the same time, because there is a finite transmission of the mirrors, part of the signal gets lost each round-trip (from each mirror). If the parametric gain is equal to the loss, then the signal can reproduce itself every round-trip, giving a stable oscillation. When this happens, we call the resulting device a *parametric oscillator*. The pump power needed for the gain to *equal* the loss is called the *threshold pump power*. When the pump power exceeds the threshold value, the gain at first exceeds the loss and the signal builds up (as does potentially the idler field if the cavity is doubly-resonant). However, once these fields start to build up appreciably, we can no longer take the pump to be un-depleted. As the pump starts to deplete, the available gain decreases until the gain is equal to the round-trip loss, leading to stable oscillation. This is the mechanism of *gain saturation*.

If you know a little bit about conventional lasers based on the inversion of a gain medium, the story is in fact somewhat similar. Once we pump the gain medium sufficiently to achieve sufficient population inversion, the small-signal gain is greater than the round-trip loss and the field builds up until the population inversion depletes due to strong stimulated emission. At equilibrium, the depleted or saturated gain is equal to the loss. Of course, the major difference with the parametric oscillator is that there is no physical gain medium that we invert with a source of energy! Here, the $\chi^{(2)}$ medium mediates energy transfer between different modes of the electromagnetic field.

In what follows, we will develop a theory of the threshold of the parametric oscillator in the singly- and doubly-resonant regimes. Then, we will work out a theory of saturation. Let us study the behavior of a signal and idler field $A_s, A_i$ that pass through a $\chi^{(2)}$ crystal which also has a pump field $A_p$. We will consider case of phase-matching for simplicity since this leads to the lowest threshold pump power.

If the pump-field is non-depleted, then a small extension of the previous section would

give (mapping $A_1, A_2, A_3 \to A_s, A_i, A_p$):

$$A_s(z) = \left[A_s(0)\cosh(\lambda z) + \frac{2i\omega_s^2 d_{\text{eff}}}{c^2 k_s \lambda} A_p A_i^*(0)\sinh(\lambda z)\right]$$
$$A_i^*(z) = \left[A_i^*(0)\cosh(\lambda z) - \frac{2i\omega_i^2 d_{\text{eff}}}{c^2 k_i \lambda} A_p^* A_s(0)\sinh(\lambda z)\right], \tag{2.5.1}$$

where $\lambda^2 = \frac{4\omega_1^2\omega_2^2 d_{\text{eff}}^2}{k_1 k_2 c^4}|A_3|^2$.

After passing through the crystal, they propagate to the mirror, and reflect off, each picking up complex amplitudes $-r_{s,1}$ and $-r_{i,1}$ respectively[17]. They then propagate backwards. We assume that there may be absorption losses $\alpha_{s,i}$ per unit length associated with propagation in the cavity. When they propagate backwards through the crystal, there is no gain associated with that (reader: why?). They then bounce off the second mirrors, picking up $-r_{s,2}$ and $-r_{i,2}$, completing their round trip. The electric fields therefore map to:

$$E_s(0) \to r_{s,1}r_{s,2}e^{2ik_s d}e^{-\alpha_s d}\left[A_s(0)\cosh(\lambda L) + \frac{2i\omega_s^2 d_{\text{eff}}}{c^2 k_s \lambda} A_p A_i^*(0)\sinh(\lambda L)\right]$$
$$E_i^*(0) \to r_{i,1}r_{i,2}e^{2ik_i d}e^{-\alpha_i d}\left[A_i^*(0)\cosh(\lambda L) - \frac{2i\omega_i^2 d_{\text{eff}}}{c^2 k_i \lambda} A_p^* A_s(0)\sinh(\lambda L)\right], \tag{2.5.2}$$

where $d$ is the cavity length and $L$ is the crystal length[18].

Let us define $\ell_s = 1 - r_{s,1}r_{s,2}e^{-\alpha_s d}, \ell_i = 1 - r_{i,1}r_{i,2}e^{-\alpha_i d}$ as the amplitude losses associated with round trip propagation. The threshold condition is that the fields reproduce themselves after this round trip. This gives the conditions

$$\cosh \lambda L = \left[1 + \frac{\ell_i \ell_s}{2 - \ell_i - \ell_s}\right]. \tag{2.5.3}$$

Typically, the gain per pass is low and we may approximate the left-hand side by $1 + \frac{1}{2}(\lambda L)^2$ giving

$$(\lambda L)^2 \approx \frac{2\ell_i \ell_s}{2 - \ell_i - \ell_s}. \tag{2.5.4}$$

In the doubly-resonant case, both losses are small compared to one and we can further simplify to

$$(\lambda L)^2 \approx \ell_i \ell_s \tag{2.5.5}$$

as the resonant condition.

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[17] I am using the convention $\begin{pmatrix} -r & it \\ it & -r \end{pmatrix}$ for the scattering matrix. This is the convention used for example in Waves and Fields in Optoelectronics by Haus. The results of course do not change with a different convention.

[18] You'll notice that I went from envelope to field. This is because in principle, the wavevector can shift due to the mirrors and the crystal, changing the resonance condition for the cavity. We will ignore this effect anyway as it is not important for the gain threshold at this level of treatment, but it is important to know that this is an effect that can occur. We will assume that $k_s, k_i$ correspond to resonances of the cavity so that $2k_i d, 2k_s d \pmod{2\pi} = 0$. This assumes that the $k$s that we are using take into account the fact that the refractive index is a weighted average of the crystal refractive index and the air/vacuum surrounding it.

In the singly-resonant case, we can take $A_i(0) = 0$ and it is straightforward to see that

$$1 = (1 - \ell_s)\cosh\lambda L \implies (\lambda L)^2 = 2\ell_s. \tag{2.5.6}$$

We see then that in the doubly-resonant case, the threshold is lower by a factor of $\sqrt{\ell_i/2}$, and so it is in principle easier to get a doubly-resonant oscillator to oscillate.

However, this comes at a cost. In a real cavity there are many modes with low loss within the gain bandwidth (defined as the range of frequencies such that $\Delta kL < 1$). In the singly-resonant case, the signal mode that oscillates is typically the one with the largest gain. The idler mode automatically satisfies $\omega_i = \omega_p - \omega_s$ and never builds up to an appreciable amplitude so there are no real constraints on the idler (it comes along for free).

In the doubly-resonant case, we want both signal and idler to build up: this means there need to be resonance modes at frequencies $\omega_s$ and $\omega_i$ such that $\omega_s + \omega_i = \omega_p$. In general, this constraint is hard to satisfy with multiple pairs of $\omega_s, \omega_i$ and there will typically only be one pair that satisfies the criterion[19]. As a result of this, the primary determinant of which modes oscillate is less which mode has the highest gain, but which modes can even satisfy the energy-conservation requirement $\omega_s + \omega_i = \omega_p$. Because of the low threshold associated with the doubly-resonant case, the reduced gain by being somewhat phase-mismatched is less problematic. However, what is a problem in the doubly resonant case is that if the cavity properties *change* due to fluctuations in the cavity length of a mechanical or thermal nature, or fluctuations in the pump frequency itself, the cavity frequencies will change. If there is a new pair of signal and idler frequencies $\omega_s', \omega_i'$ such that $\omega_s' + \omega_i' = \omega_p$, it in general will not be next to $\omega_s, \omega_i$ due to the unequal cavity mode spacings - leading to mode jumping or hopping, which is undesirable. The doubly resonant geometry amplifies frequency noise [20].

### 2.5.1 Track 2: Above-threshold behavior of the parametric oscillator

This section is focused on a detailed treatment of the parametric oscillator. This material is labeled Track 2 as it is more detailed than we normally are in this class. For even more information on this important topic, see Harris, Stephen E. "Tunable optical parametric oscillators." Proceedings of the IEEE 57.12 (1969): 2096-2113. In what follows, we will deal only with the singly resonant case as such systems are notably more stable than their

[19] I have assumed the cavity modes are not equally spaced, or that the signal and idler modes have incommensurate mode spacings (free-spectral ranges). In practice, modes are unequally spaced due to the frequency dependence of the refractive index $n(\omega)$ making it such that the frequency spacing between modes satisfies $\omega_m = \frac{ck_m}{n(\omega_m)}$ where $k_m = m\pi/L$. If we take $m \to m+1$ then we get $\Delta\omega_m \frac{d(n\omega)}{d\omega}\Big|_{\omega_m} = \pi c/L \implies \Delta\omega_m = (\pi c/L)/\frac{d(n\omega)}{d\omega}\Big|_{\omega_m}$.

[20] Suppose that the pump-frequency varies by $\delta\omega_p$. Then of course, by energy conservation $\delta\omega_p = \delta\omega_s + \delta\omega_i$. We expect that the new signal and idler frequency will, at threshold, have the same gain as the original signal and idler frequency. Why? Because, at threshold, the gain must balance the loss and the loss does not change much with frequency. Since the gain is largely determined by the phase-mismatch, we can enforce that $\delta(\Delta k) = \frac{\partial k_i}{\partial \omega_i}\delta\omega_i + \frac{\partial k_s}{\partial \omega_s}\delta\omega_s - \frac{\partial k_p}{\partial \omega_p}\delta\omega_p$. Using $\delta\omega_i = \delta\omega_p - \delta\omega_s$ and $\frac{\partial k}{\partial \omega} = \frac{n_g}{c}$ with $n_g$ the group index derived in the previous footnote, one finds: $\delta\omega_s = \frac{n_{g,p} - n_{g,i}}{n_{g,s} - n_{g,i}}\delta\omega_p$. This result among other things tells us that an operator operated near degeneracy (signal and idler frequencies nearly equal) leads to a huge linewidth. This result is derived in Kovrigin, A., and R. Byer. "Stability factor for optical parametric oscillators." IEEE Journal of Quantum Electronics 5.7 (1969): 384-385 - although it is brief!

doubly-resonant counterparts. Assume that the OPO is resonant for the signal: then we expect the idler field to be small. The idea is that the signal builds up in the resonator until there's sufficient gain to build up a significant signal and idler field. Further, we assume, as before, that the OPO is not resonant for the pump. Therefore, the pump field at the entrance facet of the crystal can be taken as a constant equal to $A_p(0)$. The coupled amplitude equations are exactly the equations of difference-frequency generation.

Let us analyze the properties of the signal field assuming that the system is oscillating in a steady-state. Then, assuming that the change in the signal field is small as it passes through the crystal, we can take its magnitude at the entrance to the crystal as a known value $A_s$ and ask about the dynamics of the pump and idler fields. The result after a length $L$ of propagation in the crystal is, assuming phase-matching $\Delta k = 0$[21]

$$\begin{aligned} A_p(L) &= A_p(0)\cos(\lambda L) \\ A_i(L) &= \frac{2id_{\text{eff}}\omega_i}{n_i c\lambda} A_s^* A_p(0)\sin(\lambda L). \end{aligned} \tag{2.5.7}$$

We can use this to then calculate the change in the signal itself using

$$\partial_z A_s = \frac{2i\omega_s d_{\text{eff}}}{n_s c} A_p(z)A_i^*(z) = \frac{2\omega_i\omega_s d_{\text{eff}}^2}{n_i n_s c^2\lambda}|A_p(0)|^2 A_s \sin(2\lambda z) \tag{2.5.8}$$

Now we will integrate this equation[22], yielding

$$A_s(L) = \exp\left[\frac{2\omega_i\omega_s d_{\text{eff}}^2}{n_i n_s c^2}|A_p(0)|^2\frac{\sin^2\lambda L}{\lambda^2}\right] A_s(0). \tag{2.5.9}$$

The overall evolution of the signal field after a round trip involves an additional loss such that after a round trip:

$$A_s \to (1-\ell_s)\exp\left[\frac{2\omega_i\omega_s d_{\text{eff}}^2}{n_i n_s c^2}|A_p(0)|^2\frac{\sin^2\lambda L}{\lambda^2}\right] A_s(0). \tag{2.5.10}$$

We have neglected here the propagation phase assuming $2k_s d \pmod{2\pi} = 0$. The oscillation condition is, assuming weak gain per pass, such that the argument of the exponent can be treated as small:

$$\frac{2\omega_i\omega_s d_{\text{eff}}^2}{n_i n_s c^2}|A_p(0)|^2\frac{\sin^2\lambda L}{\lambda^2} = \ell_s \tag{2.5.11}$$

This is the condition that amplitude gain equals amplitude loss. Recall that in this expression $\lambda = \frac{2d_{\text{eff}}}{c}\sqrt{\frac{\omega_p\omega_i}{n_p n_i}}|A_s|$.

Just above threshold, when the gain is just barely enough to match the loss (in the absence of depletion effects), we expect the steady state signal field to be small (and to go to zero exactly at threshold). In that case, our equilibrium condition is

$$\frac{2\omega_i\omega_s d_{\text{eff}}^2}{n_i n_s c^2}|A_p(0)|^2 L^2 = \ell_s \tag{2.5.12}$$

[21] Recall from the discussion above that for the singly-resonant oscillator, it likes to oscillate at a signal mode where the gain is highest.

[22] This may look fishy since in deriving the pump and idler fields, we said that $A_s$ is constant over the crystal. However, its amplitude changes only weakly over the crystal and so there is little approximation error in the pump and idler fields themselves!

which is equivalent to the singly-resonant threshold condition introduced in the previous section. Above threshold, as we increase the pump power, the only way this equality is satisfied is if $\lambda \neq 0$ corresponding to a finite value of the signal field. The signal field is determined as a function of the pump power by the solution of this equation! The origin of this reduction of gain at finite signal field is depletion of the pump. We see that if the $\lambda L$ becomes appreciable, then $A_p(L) = A_p(0)\cos(\lambda L)$ becomes smaller than $A_p(0)$ (hence depletion). We have thus worked out an elementary treatment of the above threshold behavior of the parametric oscillator.

To conclude this analysis, we consider the question of how much energy is extracted from the *idler*. This can be straightforwardly calculated: the intensity of the idler generated is

$$I_i = 2\epsilon_0 n_i c|A_i|^2 = \frac{4\epsilon_0 n_s c\omega_i}{\omega_s}\ell_s|A_s|^2. \tag{2.5.13}$$

The intensity of the signal exiting the cavity is the sum of the intensities exiting either side of the cavity and so

$$I_s = 2\ell_s \times 2\epsilon_0 n_s c|A_s|^2, \tag{2.5.14}$$

assuming that all losses from the cavity go into the beam exiting the cavity. This implies that

$$I_i = \frac{\omega_i}{\omega_s}I_s. \tag{2.5.15}$$

At the same time, we also know from the Manley-Rowe relation that every idler photon generated corresponds to the removal of a photon from the pump, and so we have that

$$\frac{I_p(L)}{\omega_p} - \frac{I_i(L)}{\omega_i} = \frac{I_p(0)}{\omega_p}. \tag{2.5.16}$$

If the pump power is chosen such that it is fully depleted at $z = L$ then $\frac{\omega_i}{\omega_p}I_p(0)$ is the maximum power that can be in the idler.

## 2.6 Electro-optic effect

The last topic that we'll discuss in our tour of effects mediated by $\chi^{(2)}$ is the electro-optic effect, and in particular, the change in the refractive index of light by static voltages. This is called the *electro-optic* effect, and it allows us to use voltages to change the phase of light accumulated by light of different orthogonal polarizations, which can be used as the basis for *electro-optic modulators*.

The modification of the refractive index by a static voltage can be understood readily in the case of an instantaneous $\chi^{(2)}$ medium, where the polarization is given by

$$P_i^{(2)}(t) = 2\epsilon_0 d_{\mathrm{ijk}} E_j(t) E_k(t). \tag{2.6.1}$$

If we consider a field which is a superposition of a DC field and a monochromatic field,

$$E = E_{\mathrm{DC}} + E_0 e^{-i\omega t} + \mathrm{c.c}, \tag{2.6.2}$$

then we see that there is a contribution at the same frequency at the monochromatic field to the polarization:

$$4\epsilon_0 d_{\mathrm{ijk}} E_{\mathrm{DC},k} E_{0,j} e^{-i\omega t}, \tag{2.6.3}$$

which looks like a change in the susceptibility [23]. Notice however that the change in the susceptibility

$$\delta\chi^{(1)}_{ij} = 4 d_{ijk} E_{\mathrm{DC},k} \tag{2.6.4}$$

is tensorial.

In the literature on the electro-optic effect, it is conventional to talk about the change in the inverse permittivity rather than the permittivity itself. The inverse of the relative permittivity $\boldsymbol{\epsilon}$ is denoted $\boldsymbol{\eta}$ and its changes are related to changes in $\boldsymbol{\epsilon}$ by [24]

$$\delta\boldsymbol{\eta} = -\boldsymbol{\eta}\delta\boldsymbol{\epsilon}\boldsymbol{\eta}. \tag{2.6.5}$$

Using $\delta\chi = \delta\epsilon$ we may write

$$\delta\eta_{ij} = r_{ijk} E_{\mathrm{DC},k}, \tag{2.6.6}$$

where

$$r_{ijk} = -4\epsilon_0 \eta_{ia}\eta_{bj} d_{abk}. \tag{2.6.7}$$

Similar to the $d$ matrix, there is a contracted notation for $r$ which follows from the symmetry of $\eta$ and full permutation symmetry of $d$. In particular, we can interchange the indices $i, j$ meaning that there are only six independent pairs of $(i, j)$. The mapping to contracted notation is $(1,1) \to 1, (2,2) \to 2, (3,3) \to 3, (2,3)/(3,2) \to 4, (3,1)/(1,3) \to 5, (1,2)/(2,1) \to 6$. The $r$ matrix is usually represented as

$$r = \begin{pmatrix} r_{11} & r_{12} & r_{13} \\ r_{21} & r_{22} & r_{23} \\ r_{31} & r_{32} & r_{33} \\ r_{41} & r_{42} & r_{43} \\ r_{51} & r_{52} & r_{53} \\ r_{61} & r_{62} & r_{63} \end{pmatrix}. \tag{2.6.8}$$

The nonzero elements are simply related to the nonzero elements of $\eta$ and $d$ through the relation $r_{ijk} = -4\epsilon_0 \eta_{ia}\eta_{bj} d_{abk}$.

Let us now explore the ramifications of the electro-optic effect. Consider for example a crystal like KDP of the $\bar{4}2m$ crystal class. It has three non-zero $r$ elements: $r_{41} = r_{52} = 8.77$ pm/V and $r_{63} = 10.5$ pm/V. Suppose that the crystal is oriented such that light propagates along the $z$ direction, parallel to one principal axis of KDP. The other two crystal axes are orthogonal to $z$. The c-axis of the crystal is taken to be in the $z$ direction. Suppose now that

[23] I have used the fact that for an instantaneous lossless medium, I can interchange indices of $d$ freely.

[24] The result below can be found as follows. Consider $(A + \delta A)^{-1}$ where $A, \delta A$ are matrices. Their inverse corresponds to solving $(A + \delta A)x = b$. Suppose that when $\delta A = 0$, $x_0 = A^{-1}b$. When $\delta A \neq 0$ but is small, we can seek a perturbative solution of the form $(A+\delta A)(x_0+\delta x) \approx Ax_0 + A\delta x + \delta A x_0 = b \implies \delta x = -A^{-1}\delta A A^{-1} b$. Therefore if $x = x_0 + \delta x \approx A^{-1} - A^{-1}\delta A A^{-1} b$, we may say that $\delta A^{-1} = A^{-1}\delta A A^{-1}$.

we apply a voltage along the $z$ axis ($\mathbf{E}_{\rm DC} = E_z\hat{z}$ with $E_z = -(V/L)$ where $V$ is the voltage difference and $L$ is the length of crystal.). The change in the inverse permittivity is then

$$\delta\eta_{ij} = \begin{pmatrix} 0 & \Delta & 0 \\ \Delta & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{2.6.9}$$

where

$$\Delta = r_{63}E_z. \tag{2.6.10}$$

This changes the principal axes in a meaningful way even for infinitesimal $E_z$. Of course $z$ is still a principal axis of the system. Meanwhile in the $xy$-plane, the principal axes of $\eta$ are found by diagonalizing

$$\eta_o I + \begin{pmatrix} 0 & \Delta \\ \Delta & 0 \end{pmatrix}, \tag{2.6.11}$$

which has eigenvalues and corresponding eigenvalues

$$\eta_o + r_{63}E_z, \eta_o - r_{63}E_z \leftrightarrow \hat{u}_1 \equiv \frac{1}{\sqrt{2}}\begin{pmatrix} 1 \\ 1 \end{pmatrix}, \hat{u_2} \equiv \frac{1}{\sqrt{2}}\begin{pmatrix} 1 \\ -1 \end{pmatrix} \tag{2.6.12}$$

We may map these changes in $\eta$ to changes in the principal values of the index of refraction by using

$$\delta\eta_o = -\frac{2\delta n}{n^3}. \tag{2.6.13}$$

Therefore, for the direction 45 degrees counterclockwise to the x-axis, the index is $n_o - \frac{1}{2}n_o^3 r_{63}E_z$ (and a wave propagates faster, making the x-axis the "fast axis"), while for the direction 45 degrees counterclockwise to the y-axis, the index is $n_o + \frac{1}{2}n_o^3 r_{63}E_z$ (making the y-axis the slow-axis)[25]

Therefore, using the electro-optic effect, we can induce birefringence for light polarized in different directions in the $xy$ plane, inducing different phase shifts for light polarized along orthogonal directions. Using this, we can construct a powerful device called an electro-optic modulator that lets us impart a voltage-dependent change in the amplitude or phase of light.

### 2.6.1 Electro-optic modulators

Consider what happens when we send light through a polarizer oriented along the $x$ direction, and then send it into a KDP crystal with a z-oriented electric field. The initial complex field

$$E = E_0\hat{x} = E_0\frac{\hat{u}_1 + \hat{u}_2}{\sqrt{2}} \tag{2.6.14}$$

will, after a length $L$ evolve into

$$E(L) = E_0 e^{i\phi_0}\frac{\hat{u}_1 + \hat{u}_2 e^{i\Gamma}}{\sqrt{2}}, \tag{2.6.15}$$

[25] Your instructor strongly believes that Boyd has the wrong sign here. The sign your instructor arrived at is the same as in Haus ("Waves and Fields in Optoelectronics") and Yariv & Yeh ("Optical Electronics in Modern Communications.")

where $\phi_0 = n_0 \frac{\omega L}{c} - \frac{1}{2} n_0^3 r_{63} E_z \frac{\omega L}{c}$ and $\Gamma$ the *retardance* is given by

$$\Gamma = n_0^3 r_{63} E_z \frac{\omega L}{c} = \frac{2\pi}{\lambda} n_0^3 r_{63} E_z L = \frac{2\pi}{\lambda} n_0^3 r_{63} |V|. \tag{2.6.16}$$

As a natural result of this birefringence, the components of the polarization along different axes achieve different rotations. Now, if we go back into the $xy$ coordinate system, we get:

$$E_x(L) = E_0 e^{i\phi_0} \frac{1 + e^{i\Gamma}}{2}, \tag{2.6.17}$$

and

$$E_y(L) = E_0 e^{i\phi_0} \frac{1 - e^{i\Gamma}}{2}. \tag{2.6.18}$$

we see that the polarization is rotated. To get at the rotation angle, suppose we put a polarizer along the x-axis. The resulting intensity, relative to the incident intensity $I_0 \sim |E_0|^2$ would be

$$I = I_0 \cos^2(\Gamma/2). \tag{2.6.19}$$

Comparing this to Malus' law, we see that the polarization rotation angle is $\Gamma/2$.

The intensity modulator works by terminating the propagation chain with the polarization in the $y$ direction rather than $x$, in which the output intensity is

$$I = I_0 \sin^2(\Gamma/2). \tag{2.6.20}$$

The principle of the intensity modulator is that changing an applied voltage changes the intensity of light, and if we modulate the voltage in time, we can modulate the intensity in time. For example by having a square voltage wave, we can create a square wave of light intensity that serves as optical bits transmitting "0"s or "1"s. Hence, such intensity modulators are of great importance in optical communications.

In the intensity modulator, it is typically important to operate at a point where a smaller change in the voltage leads to a larger change in the intensity. That requires operating around the voltage such that $\Gamma = \pi/2$, which corresponds to the voltage

$$\frac{4}{\lambda} n_0^3 r_{63} |V| = 1 \implies |V| = \frac{\lambda}{4 n_0^3 r_{63}}. \tag{2.6.21}$$

This is related to a figure of merit called the *half-wave voltage* which is the voltage needed to get a $\pi$ phase shift (a half-wavelength's worth of phase shift). That voltage is simply

$$V_{\lambda/2} = \frac{\lambda}{2 n_0^3 r_{63}}. \tag{2.6.22}$$

Typical magnitudes of this voltage are on the order of 10 kV and require high-voltage supplies!

Finally, we mention that if we send light polarized along one of the new principal axes, say $\hat{u}_1$, then light just experiences a phase shift proportional to the voltage. By varying that voltage in time, we modulate the *phase* directly, which is important for communication based on the optical phase of light.

# Chapter 3

# Effects arising from the third-order nonlinear susceptibility

In this chapter, we will study a handful of important effects arising $\chi^{(3)}$. In the previous chapter, we focused on various frequency-conversion effects such as harmonic generation, sum- and difference-frequency generation, and electro-optic modulation. While many of these effects can also be realized by $\chi^{(3)}$, we will focus on the most unique effects. At the core of these effects is the idea of an *intensity-dependent index of refraction* [1].

## 3.1 Nonlinear index of refraction

Consider a third-order nonlinear medium. The relationship between electric field and polarization, in frequency-domain, can be written as

$$P_i(\mathbf{r},\omega) = \int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi}\frac{d\omega_3}{2\pi}\delta(\omega-\omega_\sigma)\chi^{(3)}(\omega_\sigma;\omega_1,\omega_2,\omega_3)E_j(\omega_1)E_k(\omega_2)E_l(\omega_3). \tag{3.1.1}$$

Needless to say, this expression is quite complicated due to the 81 different combination of indices (in three dimensions) for a given set of frequency labels. Symmetries simplify this behavior, but only to an extent. We will, therefore, consider two types of simplifications for our analysis, but do keep in mind they do not always hold. We will start with the extraordinarily simple (but one of the most practically useful) cases of a lossless material, in which we may write the time-domain relation

$$P_i(\mathbf{r},\omega) = \epsilon_0\chi^{(3)}_{ijkl}E_j(\mathbf{r},t)E_k(\mathbf{r},t)E_l(\mathbf{r},t). \tag{3.1.2}$$

Let us now consider what happens when we have a monochromatic field of the form

$$E(\mathbf{r},t) = E(\mathbf{r})e^{-i\omega t} + \text{c.c.} \tag{3.1.3}$$

There are polarization terms at $\pm 3\omega$ and $\pm\omega$. The $\pm 3\omega$ terms lead to third-harmonic generation, which typically is hard to phase-match for reasons discussed in the previous unit.

[1] These effects are also realizable via second-order nonlinear media, where under some circumstances $\chi^{(2)}$ can act as an effective $\chi^{(3)}$ but there are provisos here and the effects we will describe are much more commonly explored in the third-order case.

The $\pm\omega$ polarization terms however lead to an effective correction to the refractive index that is always relevant. The corresponding polarization is

$$P_i(\mathbf{r},\omega) = \epsilon_0 \chi^{(3)}_{ijkl} \left( E_j(\mathbf{r}) E_k(\mathbf{r}) E^*_l(\mathbf{r}) + E_j(\mathbf{r}) E^*_k(\mathbf{r}) E_l(\mathbf{r}) + E^*_j(\mathbf{r}) E_k(\mathbf{r}) E_l(\mathbf{r}) \right). \tag{3.1.4}$$

In a lossless material, we may use Kleinman symmetry to write

$$P_i(\mathbf{r},\omega) = \epsilon_0 \left( \chi^{(3)}_{ijkl} E_j(\mathbf{r}) E_k(\mathbf{r}) E^*_l(\mathbf{r}) + \chi^{(3)}_{ijkl} E_j(\mathbf{r}) E^*_k(\mathbf{r}) E_l(\mathbf{r}) + \chi^{(3)}_{ijkl} E^*_k(\mathbf{r}) E_j(\mathbf{r}) E_l(\mathbf{r}) \right). \tag{3.1.5}$$

This can be written as an effective anisotropic index of refraction, via

$$P_i(\mathbf{r},\omega) = \epsilon_0 \left( \chi^{(3)}_{ijkl} E_j(\mathbf{r}) E_k(\mathbf{r}) E^*_l(\mathbf{r}) + 2\chi^{(3)}_{ijkl} E_j(\mathbf{r}) E^*_k(\mathbf{r}) E_l(\mathbf{r}) \right) \equiv \epsilon_0 \chi_{\text{eff},ij}(\mathbf{r}) E_j(\mathbf{r}), \tag{3.1.6}$$

with

$$\chi_{\text{eff},ij}(\mathbf{r}) = \chi^{(3)}_{ijkl} E_k(\mathbf{r}) E^*_l(\mathbf{r}) + 2\chi^{(3)}_{ijkl} E^*_k(\mathbf{r}) E_l(\mathbf{r}) = 3\chi^{(3)}_{ijkl} E^*_k(\mathbf{r}) E_l(\mathbf{r}). \tag{3.1.7}$$

The exact anisotropy depends on the nonzero indices of the third-order susceptibility, but we can see already at this stage schematically that the polarization at the same frequency acts almost like an intensity-dependent index of refraction. To simplify further, we will make use of symmetry.

### 3.1.1 Effect of symmetries

Symmetries strongly reduce the number of independent components. A detailed group theoretic analysis of which crystal classes have how many independent elements is beyond the scope of this work, but we will consider one example, which is the isotropic case. For an isotropic system, the number of independent elements is one (rather than 81!). This can be understood using the quartic oscillator as an example. We derived the quartic oscillator nonlinear susceptibility assuming an isotropic potential. The result was:

$$\chi^{(3)}_{ijkl}(\omega_\sigma;\omega_1,\omega_2,\omega_3) = -\frac{nq^4}{3m^3\epsilon_0} \frac{\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)}. \tag{3.1.8}$$

Clearly all elements are proportional to $-\frac{nq^4}{3m^3\epsilon_0}\frac{1}{D(\omega_\sigma)D(\omega_1)D(\omega_2)D(\omega_3)}$. In fact, it turns out that in an isotropic system, the susceptibility *must* have the tensor structure of the quartic oscillator we analyzed. We may therefore write a generic isotropic lossless third-order susceptibility as

$$\chi^{(3)}_{ijkl} = \frac{1}{3}\chi^{(3)}(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}), \tag{3.1.9}$$

where $\chi^{(3)}$ corresponds to an element such as $\chi^{(3)}_{1111}$. Plugging this relationship into the nonlinear polarization at frequency $\omega$, we get

$$P_i(\mathbf{r},\omega) = \epsilon_0\chi^{(3)}(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) E^*_k(\mathbf{r}) E_l(\mathbf{r}) E_j(\mathbf{r}), \tag{3.1.10}$$

which may be written as

$$\mathbf{P}(\mathbf{r},\omega) = \epsilon_0\chi^{(3)} \left( 2(\mathbf{E}(\mathbf{r})\cdot\mathbf{E}^*(\mathbf{r}))\mathbf{E}(\mathbf{r}) + (\mathbf{E}(\mathbf{r})\cdot\mathbf{E}(\mathbf{r}))\mathbf{E}^*(\mathbf{r}) \right). \tag{3.1.11}$$

A term like this acts as an effective polarization rotation, which you will explore on the problem set.

Let us however now consider the very simple case in which the field is polarized in a single direction, e.g., $\hat{x}$, so that we get

$$P(\mathbf{r},\omega) = 3\epsilon_0\chi^{(3)}|E(\mathbf{r})|^2E(\mathbf{r}), \tag{3.1.12}$$

where the scalarized form of $\mathbf{P},\mathbf{E}$ should be understood e.g., as $\hat{x}\cdot\mathbf{P}$. Let us now explore the effective index of refraction for an $x$-polarized wave as follows. The scalar Helmholtz equation in the presence of a source is given by

$$\left(\nabla^2 + \left(\frac{n_0\omega}{c}\right)^2\right)E(\mathbf{r}) = -\mu_0\omega^2 P_{\rm NL} = -3\frac{\omega^2}{c^2}\chi^{(3)}|E(\mathbf{r})|^2E(\mathbf{r}), \tag{3.1.13}$$

where $n_0 = n(\omega)$ is the linear index of refraction at frequency $\omega$. Clearly, by moving the right-hand side over to the left-hand side, we have

$$\left(\nabla^2 + \left(\frac{n(I)\omega}{c}\right)^2\right)E(\mathbf{r}), \tag{3.1.14}$$

where

$$n(I)^2 = n_0^2 + 3\chi^{(3)}|E(\mathbf{r})|^2 \implies n(I) = n_0 + \frac{3\chi^{(3)}|E(\mathbf{r})|^2}{2n_0} = n_0 + \frac{3\chi^{(3)}}{4n_0^2\epsilon_0 c}I. \tag{3.1.15}$$

This is typically expressed as

$$n(I) = n_0 + n_2 I, \tag{3.1.16}$$

where

$$n_2 = \frac{3\chi^{(3)}}{4n_0^2\epsilon_0 c}. \tag{3.1.17}$$

The quantity $n_2$ is called the nonlinear index of refraction and has units of inverse intensity. In MKS units, there are $\mathrm{m}^2/\mathrm{W}$.

## 3.2 Effect of the intensity-dependent refractive index on short pulses

The third-order nonlinear contribution is much weaker than the second-order nonlinear contribution (in a centrosymmetry breaking material) in the following sense: we know that the polarization is a perturbation expansion and that the contribution of the third-order term is like $P^{(3)} \sim P^{(1)}(E/E_0)^2 \sim P^{(2)}(E/E_0)$ and so for $E \ll E_0 \sim 0.1\mathrm{V/pm}$, the third-order contribution to the polarization is very small in magnitude. Said in terms of the nonlinear index of refraction, consider light with an intensity of 1 $\mathrm{GW/cm}^2$. In a material like amorphous silica, where $n_2 \sim 10^{-20}\mathrm{m}^2/\mathrm{W}$, the corresponding change in the refractive index is only $10^{-7}$ which is very small. It is possible to have small changes like this lead to large effects, particularly when the light propagates over long distances. To achieve these intensities in the first place, it helps to have both well-focused light (confined to a small lateral dimension)

and to compress the energy of the light into a short pulse, which enhances the instantaneous electric field. Since we are considering instantaneous materials, their polarization responds to the instantaneous field and intensity. This brings us to an analysis of how short pulses propagate in third-order nonlinear media.

Let us consider a simple case of short pulses propagating in $\chi^{(3)}$ media in which the light can be described effectively as one-dimensional, propagating in a single direction $z$ with fixed polarization. In that case, the relevant self-polarization terms are along the same direction as the electric field and we may consider a nonlinear scalar wave treatment. Although this treatment sounds artificial, it well-describes a situation in which light is confined to propagate along an optical waveguide. One of the most technologically relevant examples of an optical waveguide is an optical fiber. An example of an optical fiber includes a high-index "core" region of some radius surrounded by a lower index cladding region. The difference in index can be small, on the order of 0.02. We know from Maxwell's equations that a region of high-index acts as a region of negative potential [2]. This core-cladding difference is sufficient to trap a small number of bound states. Since they are bound transversally, their transverse dynamics are effectly frozen and we can focus purely on field variations in the $z$ direction. The result of this treatment is effectively to recover the one-dimensional pulse propagation equation that we derived on the homework. To see this, let us start by writing the full spatiotemporal electric field, corresponding to a particular transverse mode $u(\mathbf{r})$ polarized in the $\hat{x}$ direction. This is possible to achieve by using polarization-maintaining fibers that exploit anisotropy to break the degeneracy between $\hat{x}$ and $\hat{y}$ polarized modes. The field may be written as [3]

$$\mathbf{E}(\mathbf{r},t) = \hat{x}u_k(\mathbf{r})A(z,t)e^{ikz-i\omega t} + \text{c.c.} \tag{3.2.1}$$

The corresponding field, in the frequency domain, could be expressed as:

$$\mathbf{E}(\mathbf{r},\omega) = \hat{x}u_k(\mathbf{r})A(z,\omega)e^{ikz}. \tag{3.2.2}$$

Let us consider the Maxwell equations in the absence of nonlinear polarization for the time being. The source-free frequency domain Maxwell equation is

$$\left(\nabla^2 + n^2(\mathbf{r})\frac{\omega^2}{c^2}\right)\mathbf{E}(\mathbf{r},\omega) = 0 \tag{3.2.3}$$

The left-hand side simplifies as follows. The Maxwell eigenmode $u_k(\mathbf{r})$ satisfies

$$\nabla_\perp^2 u_k(\mathbf{r}) + \left(n^2(\mathbf{r})\frac{\omega^2}{c^2} - k^2(\omega)\right)u_k(\mathbf{r}) = 0, \tag{3.2.4}$$

where $\nabla_\perp^2$ denotes the Laplacian with respect to the transverse coordinates. In writing $k(\omega)$, I've used the fact that the $\omega$ acts as an eigenvalue which is dictated by $k$ so that $\omega = \omega(k)$.

[2] As a rudimentary example, consider the Helmholtz equation for a scalar wave propagating in a variable refractive index: $\nabla^2 E(\mathbf{r},\omega) = -\frac{\omega^2}{c^2}n^2(\mathbf{r})E(\mathbf{r},\omega)$. This is formally equivalent to a Schrodinger equation with $\frac{\hbar^2}{2m} \to \frac{c^2}{\omega^2}$ and $V = -n^2$. Hence, a high index region can act as a potential well that supports confined states of light. The presence of a propagation wavevector in some direction replaces the mass term $\frac{c^2}{\omega^2}$ with $\frac{1}{\omega^2/c^2 - k^2}$.

[3] As we will see below, the eigenfunction depends on $k$.

Alternatively, if we fix $\omega$ then there is a specific $k = k(\omega)$ for which $u_k(\mathbf{r})$ has eigenfrequency $\omega$. Using this fact, we are left with the left-hand side of the Maxwell equation being

$$u_k(\mathbf{r}) \left( \frac{d^2}{dz^2} + k^2(\omega) \right) A(z,\omega) e^{ikz} = 0 \tag{3.2.5}$$

By the same set of steps that we followed in Problem Set 1 (slowly varying envelope approximation, writing $k(\omega)$ in a Taylor series), we have that

$$u_k(\mathbf{r}) e^{ikz} \left( 2ik \frac{dA}{dz} + 2k \sum_m \frac{\beta_m}{m!} (\omega - \omega_0)^m \right) A(z,\omega) = 0. \tag{3.2.6}$$

In time-domain, this may be expressed as

$$u_k(\mathbf{r}) e^{ikz - i\omega t} \left( 2ik \partial_z A(z,t) + 2k \sum_m \frac{i^m \beta_m}{m!} \partial_t^m \right) A(z,t) = 0. \tag{3.2.7}$$

Let us now consider the effect of the nonlinear polarization. Its effect is to add to the right hand side $\mu_0 \partial_t^2 P_x(\mathbf{r},t)$. This polarization may be written as

$$P_x(\mathbf{r},t) = 3\epsilon_0 \chi^{(3)} \left( u_k(\mathbf{r}) A(z,t) e^{ikz - i\omega t} + \text{c.c} \right)^3. \tag{3.2.8}$$

The only relevant parts are those which oscillate near frequency $\omega$ by construction $A(z,t)$ is considered to be an envelope field tightly centered spectrally around $\omega$. That term is

$$P_x(\mathbf{r},t) = 3\epsilon_0 \chi^{(3)} |u_k(\mathbf{r})|^2 u_k(\mathbf{r}) e^{ikz - i\omega t} |A(z,t)|^2 A(z,t). \tag{3.2.9}$$

The second time-derivative of this term, since the envelope varies in time slowly compared to the oscillation at $\omega$, is approximately $-\omega^2$ and so the resulting equation is then

$$u_k(\mathbf{r}) \left( 2ik \partial_z A(z,t) + 2k \sum_m \frac{i^m \beta_m}{m!} \partial_t^m \right) A(z,t) = -3 \frac{\omega^2}{c^2} \chi^{(3)} |u_k(\mathbf{r})|^2 u_k(\mathbf{r}) |A(z,t)|^2 A(z,t). \tag{3.2.10}$$

To complete the derivation, we note that since $u_k(\mathbf{r})$ is an orthonormal eigenmode of the Maxwell equations [4], we can project it out, writing

$$\left( \partial_z A(z,t) - i \sum_m \frac{i^m \beta_m}{m!} \partial_t^m \right) A(z,t) = \frac{3i\omega^2 \chi^{(3)}_{\text{eff}}}{2c^2 k} |A(z,t)|^2 A(z,t), \tag{3.2.11}$$

where

$$\chi^{(3)}_{\text{eff}} = \chi^{(3)} \int d\mathbf{r} |u_k(\mathbf{r})|^4. \tag{3.2.12}$$

This equation is typically re-arranged in the form:

$$\left( \partial_z + \beta_1 \partial_t \right) A(z,t) = \sum_{m=2} \frac{i^{m+1} \beta_m}{m!} \partial_t^m A(z,t) + \frac{3i\omega \chi^{(3)}_{\text{eff}}}{2cn_0} |A(z,t)|^2 A(z,t). \tag{3.2.13}$$

[4] The orthonormality condition is $\int d\mathbf{r} \epsilon(\mathbf{r}) |u_k(\mathbf{r})|^2 = 1$

We may further simplify the coefficients on the right hand side by identifying $\frac{3i\omega\chi^{(3)}_{\text{eff}}}{2cn_0} \equiv \gamma$, yielding:

$$\left(\partial_z + \beta_1\partial_t\right) A(z,t) = \sum_{m=2} \frac{i^{m+1}\beta_m}{m!}\partial_t^m A(z,t) + i\gamma|A(z,t)|^2 A(z,t). \tag{3.2.14}$$

You'll notice that we have separated the first time-derivative term from the remaining. Indeed, the two terms on the left hand side represent a "one-way" wave equation which have as a solution any function $f(t-\beta_1 z)$: i.e., a function moving at the group velocity $\beta_1$. It is conventional to go into a moving frame which removes this motion: it leads to simpler forms of the solutions. To make this transformation, take $z' = z, t' = t - \beta_1 z = t - \beta_1 z'$. The chain rule tells us then that

$$\partial_z = \partial_{z'}\frac{\partial z'}{\partial z} + \partial'_t\frac{\partial t'}{\partial z} = \partial_{z'} - \beta_1\partial_{t'} \tag{3.2.15}$$

and

$$\partial_t = \partial_{z'}\frac{\partial z'}{\partial t} + \partial'_t\frac{\partial t'}{\partial t} = \partial'_t. \tag{3.2.16}$$

Plugging these substitutions in gives us

$$\partial_{z'} A(z',t') = \sum_{m=2} \frac{i^{m+1}\beta_m}{m!}\partial_{t'}^m A(z',t') + i\gamma|A(z',t')|^2 A(z',t'). \tag{3.2.17}$$

This brings us to the final form of the so-called nonlinear short pulse propagation equation that we will use in this class. Before analyzing it, let us discuss the physical significance of this transformation. The new variable $z'$ being the same as $z$ is the physical distance along the waveguide. The new variable $t'$ is like a *retarded time* from electrodynamics. It is related to the "absolute" time by subtracting the time it takes the pulse to arrive to a particular location. That has the following physical interpretation. Let's park at some particular location in the fiber $z'$ and watch a pulse go by us. Suppose we are at a $z'$ such that it takes 1 ns for the peak of the pulse to reach that location (do not worry about dispersion and nonlinearity right now). Suppose the pulse starts propagating in the waveguide from $z' = 0$ at $t = 0$. Then at $t = 1$ ns (absolute time), at our $z' \neq 0$, we see the pulse passing by. The corresponding $t'$ for the center of the pulse is zero. The leading edge of the pulse corresponds to negative $t'$ and trailing edge to positive $t'$.

In the presence of other effects like nonlinearity and dispersion, we will see the pulse profile in time at some fixed $z$ get distorted. For example, if the pulse peak arrives at positive $t'$ then it slowed down upon its propagation. More generally, the time profile we see, parked at some $z$ is more or less the same as what happens when we take a snapshot at fixed time and look at the spatial dependence of the pulse (for short enough pulses not undergoing very rapid dynamics in $z$. That is because in the absence of other effects, the solution is $f(z,t) = f(0, t-\beta_1 z)$ which translates rigidly at the group velocity [5].

As one last important point about the moving frame. Suppose there is no higher-order dispersion $\beta_{m\geq 2} = 0$ and no nonlinearity $\gamma = 0$. Then $\partial'_z A(z',t') = 0$ and so $A(z',t') =$

[5] More explicitly, if we pursue the line or argument we above, let's fix $z$ and watch the pulse go by us in time. That is described by fixing $z$ and letting $t$ vary in $f(z,t) = f(0, t-\beta_1 z)$. If we take a snapshot in time and look at the spatial dependence of the pulse, that's described by the very same function with $t$ fixed. As you can see, these two are related by a reflection and scaling. The reflection is why the trailing edge of the pulse is positive $t'$. The scale is just a change of units from space to time.

$A(0,t') = A(0,t-\beta_1 z)$. The pulse, in this moving frame, does not evolve at all: it has the same shape as it had at $z=0$. That is the benefit of the moving frame: it moves out these trivial translational dynamics. Hopefully this discussion provides you with a lot of intuition for how $z', t'$ work. Now we will now study the consequences of this equation systematically.

In what follows, since $z' = z$ we will use $z$ instead of $z'$ for notational simplicity.

### 3.2.1 Linear dynamics

Let us start by considering the linear limit, which is obtained by setting $\gamma = 0$. Our equation is therefore

$$\partial_z A(z,t') = \sum_{m=2} \frac{i^{m+1}\beta_m}{m!} \partial_{t'}^m A(z,t'). \tag{3.2.18}$$

This is trivially solved in Fourier domain. Fourier transforming in time, we get

$$\partial_z A(z,\Omega) = \sum_{m=2} \frac{i^{m+1}\beta_m}{m!} (-i\Omega)^m A(z,\Omega) = i\beta(\Omega) A(z,\Omega), \tag{3.2.19}$$

where we have defined

$$\beta(\Omega) = \sum_{m=2}^{\infty} \frac{\beta_m}{m!} \Omega^m. \tag{3.2.20}$$

We have defined the frequency variable as $\Omega$ to remind us that this frequency describes time variations of the *envelope*. Time variations of the total field are related by a shift by $\omega$. In particular

$$E(\omega + \Omega) \sim A(\Omega). \tag{3.2.21}$$

In Fourier domain, we then see that

$$A(z,\Omega) = A(0,\Omega) e^{i\beta(\Omega) z}. \tag{3.2.22}$$

Each spectral (Fourier) component picks up a (relative) frequency dependent phase. Let's explore the consequences of this in the time-domain by considering a simple situation in which the only non-negligible dispersion is second-order, which is called group velocity dispersion (GVD). In this case, the time-domain field is

$$A(z,t') = \int \frac{d\Omega}{2\pi} e^{-i\Omega t' + \frac{1}{2} i\beta_2 \Omega^2 z} A(0,\Omega). \tag{3.2.23}$$

From this form, it is clear why $\beta_2$ represents a group velocity dispersion: each spectral component moves as a plane wave with an $\Omega$ dependent phase velocity which is, in the lab frame, $(\beta_1 + \frac{1}{2}\beta_2\Omega)^{-1}$. Alternatively, we could say that the inverse group velocity is $dk/d\omega = \beta_1 + \beta_2\Omega$. Let us consider now what happens when the initial pulse is a Gaussian, with $A(0,t) = A_0 e^{-t^2/(2\tau^2)}$ with $\tau$ the pulse duration. We solved this problem on the first problem set and we know that the solution for the time-domain field at finite $z'$ is

$$A(z,t') = A_0 \sqrt{\frac{\tau^2}{\tau^2 - i\beta_2 z}} \exp\left[ -\frac{t'^2}{2(\tau^2 - i\beta_2 z)} \right]. \tag{3.2.24}$$

The instantaneous pulse intensity [6], which is given by $I(\mathbf{r}, z, t') \sim |E(z,t')|^2 = |u_k(\mathbf{r})|^2 |A(z,t')|^2$. Typically we don't care too much about the variations in the transverse plane as we typically collect all of the light within the transverse plane, and so we average over transverse coordinates. Therefore, the instantaneous optical *power* $P(z,t') \sim |A(z,t')|^2$. Therefore, when we talk about optical power, we will use it interchangeably with $|A(z,t')|^2$ and in some cases even normalize $A$ such that its square has dimensions of power[7].

The optical power is then in the appropriate units

$$P(z,t') = P_0 \frac{1}{\sqrt{1+\frac{\beta_2^2 z^2}{\tau^4}}} \exp\left[-\frac{t^2}{\tau^2+\frac{\beta_2^2 z^2}{\tau^2}}\right]. \tag{3.2.27}$$

To simplify the interpretation, define a *dispersion length* $L_D = \tau^2/\beta_2$, which lets us write the above as

$$P(z,t') = P_0 \frac{1}{\sqrt{1+(z/L_D)^2}} \exp\left[-\frac{t^2}{\tau^2(1+(z/L_D)^2)}\right]. \tag{3.2.28}$$

This equation describes a *spreading* of the pulse and a concomitant dilution of the energy over a longer time period (causing a reduction in power). The pulse duration increases by a factor of $\sqrt{2}$ over $z = L_D$. Over a very long distance of propagation, the pulse duration eventually becomes $\tau z/L_D$ and the peak power decreases as $1/z$.

Beyond the reduction of the optical power, another subtle effect that occurs is *chirping*. If we look at a particular portion or time-segment of the pulse, we will see that depending on the sign of the dispersion, the trailing edge is redder than the leading edge or vice-versa. How do we see this? The inverse group velocity is frequency-dependent: $v_g^{-1} = \beta_1 + \beta_2\Omega$ and so for normal dispersion ($\beta_2 > 0$) the bluer parts of the pulse move slower (corresponding to a higher index of refraction, in connection to our discussion of phase-matching in the previous unit). The redder parts move faster. So the bluer parts should be at the leading edge of the pulse (negative $t'$). For anomalous dispersion $\beta_2 < 0$, the situation is reversed.

Let us define a type of windowed Fourier transform. The windowed Fourier transform of a

[6]Note that this is still time-averaged over an optical cycle which is assumed to be short relative to the envelope variations in time

[7]Note that if we elect for such a normalization, we must change the expression for $\gamma$. As we derived the equation $|A|^2$ has dimensions of $[|E|^2]L^2$ which has dimensions of $[P/\epsilon_0 c]$. To change the units of $|A|^2$ to be power, we must redefine $\gamma$ to be

$$\gamma = \frac{3i\omega\chi^{(3)}_{\text{eff}}}{2\epsilon_0 c^2 n_0} = 2n_0 n_2 \frac{\omega}{c} \int d\mathbf{r}\ |u(\mathbf{r})|^2 \tag{3.2.25}$$

. The integral has dimensions of inverse area, and is typically defined as an effective inverse area $A_{\text{eff}}^{-1}$. To match the literature, we also adjust the scale of $|A|$ by a factor of $\sqrt{2}$ so that $A \to \sqrt{2}A$. The factor of 2 can be seem as coming from enforcing that $|A|^2$ itself is the optical power. Note that any adjustment of scale (even dimensionless adjustments) requires a change in the coefficients due to the nonlinearity. This lets us write

$$\gamma = \frac{2\pi n_0 n_2}{\lambda A} \tag{3.2.26}$$

which has dimensions of $[P^{-1}]L^{-1}$, which is a standard and frequently used expression relating the nonlinear index to the coefficient in the nonlinear pulse propagation equation.

function is defined as

$$f_g(\omega, t_0) = \int_{-\infty}^{\infty} dt\ f(t)g(t - t_0)e^{i\omega t}, \tag{3.2.29}$$

where $g$ is a gating or windowing function. It defines a short interval of time over which the signal is Fourier transformed. The usual transform is obtained by $g = 1$. Let us consider what happens when the gating function is a window function which is 1 between $-\Delta t/2$ and $\Delta t/2$. In that case, the windowed Fourier transform is

$$\int_{t_0-(\Delta t)/2}^{t_0+(\Delta t)/2} dt\ f(t)e^{i\omega t}. \tag{3.2.30}$$

Let's now apply this to our dispersed Gaussian pulse. We will consider a simple but important physical limit which is the so called "far-field limit" (in analogy with diffraction theory). The far-field limit is $z \ll L_D$, so the pulse has dispersed considerably.

In this case we can write the time-domain dispersed pulse

$$A(z, t') = \sqrt{P(z, t')}\sqrt{\frac{1 + iz/L_D}{\sqrt{1 + (z/L_D)^2}}} \exp\left[-\frac{i}{2}\frac{(z/L_D)t^2}{\tau^2(1 + (z/L_D)^2)}\right], \tag{3.2.31}$$

as approximately

$$A(z, t') = \sqrt{P(z, t')} \exp\left[-\frac{iL_D t^2}{2\tau^2 z} + \frac{i\pi}{4}\right]. \tag{3.2.32}$$

Now consider the windowed Fourier transform which is proportional to

$$A(\Omega, t_0) = e^{i\frac{\pi}{4}} \int_{t_0-(\Delta t)/2}^{t_0+(\Delta t)/2} dt'\ e^{i\Omega t'} \sqrt{P(z, t')} \exp\left[-\frac{iL_D t'^2}{2\tau^2 z}\right]. \tag{3.2.33}$$

Since the exponent oscillates very rapidly for $t'$ comparable to the dispersed pulse duration, i.e, for $t' \sim \sqrt{z/L_D}\tau$, we can perform a stationary phase approximation. The stationary phase approximation tells us that the $t'$ values that determine the integral are the ones that make the derivative of the phase vanish: $i\Omega - iL_D t'/(\tau^2 z) = 0 \implies \Omega = t'/(\beta_2 z)$. Not worrying about overall factors, this gives

$$A(\Omega, t_0) \sim \sqrt{P\left(z, \beta_2 z\Omega\right)} \text{ if } \beta_2\Omega z \in [t_0 - \Delta t/2, t_0 + \Delta t/2], \text{ else } 0. \tag{3.2.34}$$

The if is because if $\Omega = t'/(\beta_2 z)$ is satisfied for a $t'$ outside of our integration window then we don't pick up the stationary phase point in the integral and we expect to get a much lower value which may be approximated as zero. That says that time and frequency get locked together: the frequency at that time slice is very concentrated around $t_0/(\beta_2 z)$. This is precisely the result we were looking for. The spectrum around time slice at $t_0$ only has a meaningful value if $\beta_2\Omega z = t_0$ (up to the width of the window function).

Of course, this could have been anticipated by our group velocity analysis! If we have a spectrum of the initial pulse which consists of temporally localized photons, and photons of

different colors move at different group velocities $v_g \approx \frac{1}{\beta_1} - \frac{\beta_2 \Omega}{\beta_1^2}$ [8], then after a long distance, photons of different colors will be separated from each other. Photons at (relative) frequency $\Omega$ will at a distance $z$ be lag the photons at $\Omega = 0$ by a distance $\delta z = \frac{\beta_2 \Omega}{\beta_1^2}(\Delta t_{\text{lab}}) = \frac{\beta_2 \Omega}{\beta_1^2}\beta_1 z$. Converting the spatial separation into a temporal separation, we get a temporal separation of $\beta_2 \Omega z$ as expected: a higher frequency component will be slower and will end up at the trailing edge of the pulse (positive $t'$). This is precisely the same type of thing that occurs via a prism or a grating except that a prism separates colors in angle, and this "time prism" separates the colors temporally (or longitudinally in space).

### 3.2.2 Nonlinear dynamics without dispersion: self-phase modulation

Let us now look at what the nonlinear terms do. To make things simple, we will consider what happens in the absence of dispersion $\beta_{m\geq 2} = 0$. In this case, we have

$$\partial_z A(z,t') = i\gamma |A(z,t')|^2 A(z,t'). \tag{3.2.35}$$

This is in fact exactly solvable. That is because the instantaneous intensity is conserved by this equation. Consider

$$\partial_z |A(z,t')|^2 = 2\text{Re } A^*(z,t')\partial_z A(z,t') = 2\text{Re } i\gamma |A(z,t')|^4 = 0. \tag{3.2.36}$$

Therefore $|A(z,t')|^2 = |A(0,t')|^2$ and

$$\partial_z A(z,t') = i\gamma |A(0,t')|^2 A(z,t'). \tag{3.2.37}$$

This can therefore be directly integrated, yielding

$$A(z,t') = \exp\left[i\gamma z |A(0,t')|^2\right] A(0,t'). \tag{3.2.38}$$

This equation has the interpretation that each time slice of the pulse (which is equivalent to points on the pulse) picks up a phase proportional to the intensity of the pulse at that time slice. This phase is called the *nonlinear* phase and is given instantaneously by

$$\phi_{\text{NL}}(z,t') = \gamma z |A(0,t')|^2. \tag{3.2.39}$$

This is, appropriately, called *self-phase modulation*. As we see, the instantaneous power does not change at all, but the spectrum does change.

Self-phase modulation also leads to a chirp. To see this, let us consider our windowed Fourier transform again. Let us assume that the nonlinear phase is sufficiently large such that we can use stationary phase techniques again to compute the windowed Fourier transform with a square windowing function. The windowed Fourier transform is equal to

$$A(\Omega, t_0) = \int_{t_0-(\Delta t)/2}^{t_0+(\Delta t)/2} dt\ e^{i\Omega t' + i\phi_{\text{NL}}(z,t')} \sqrt{P(0,t')} e^{i\phi(0,t')}, \tag{3.2.40}$$

[8] This from inverting $\beta_1 + \beta_2 \Omega$.

where we have expressed the initial pulse ($z = 0$) in polar form in anticipation of using stationary phase and we have defined the phase of the initial pulse as $\phi(0, t')$. Let us consider a purely real and thus unchirped Gaussian pulse as an example. Then the stationary phase condition is

$$\Omega = -\partial_{t'}\phi_{\mathrm{NL}}(z, t') = \gamma z|A_0|^2 \frac{2t}{\tau^2} e^{-t^2/\tau^2}. \tag{3.2.41}$$

We see in this case that the frequency at some particular time slice is redder for $t < 0$ (leading edge) and bluer for $t > 0$ (trailing edge).

### 3.2.3 Solitons

We now consider what happens when dispersion and nonlinearity are simultaneously present in our system. This is a rich regime with a lot of complicated physics, some of which remains even today to be an active form of research. There are typically very few analytical solutions in this regime. However, under specific conditions with only second-order dispersion and nonlinearity, the dynamics are analytically soluble. Although these solutions have been known for decades, small changes of the equations evade analytical solutions.

To give some intuition for the fundamental idea, we notice that self-phase modulation pushes high frequencies to $t > 0$, while group velocity dispersion with $\beta_2 > 0$ does the same thing, and the two effects reinforce each other. On the other hand, if the dispersion is *anomalous* and $\beta_2 < 0$ then the two effects could even cancel out. It turns out this cancellation is perfect and a pulse can propagate in a fully shape-preserving manner. We can get at this idea as follows. There is a way to find this shape-preserving solution, called an *soliton* exactly but it is very involved and for this course we will content ourselves with stating the solution.

The following solution is an example of a soliton.

$$A(0, t') = A_0 \mathrm{sech}(t/\tau). \tag{3.2.42}$$

Remarkably, its $z$ evolution is

$$A(0, t') = A_0 \mathrm{sech}(t/\tau) e^{\frac{1}{2} i\gamma |A_0|^2 z}. \tag{3.2.43}$$

It propagates and just picks up a nonlinear phase.

To see this, let's compute the right-hand side. Using $\mathrm{sech}(x)'' = \mathrm{sech}(x) - 2\mathrm{sech}^3(x)$, we have that the right hand side of the pulse propagation equation:

$$\partial_z A(z, t') = -i\frac{\beta_2}{2}\partial_{t'}^2 A(z, t') + i\gamma |A(z, t')|^2 A(z, t'), \tag{3.2.44}$$

which in this case is called the *nonlinear Schrodinger equation* is,

$$\left( -\frac{i\beta_2}{2\tau^2} A_0 \left( \mathrm{sech}(t'/\tau) - 2\mathrm{sech}^3(t'/\tau) \right) + i\gamma |A_0|^2 A_0 \mathrm{sech}^3(t'/\tau) \right) e^{\frac{1}{2} i\gamma |A_0|^2 z}. \tag{3.2.45}$$

While the right hand side is simply

$$\frac{1}{2} i\gamma |A_0|^2 A_0 \mathrm{sech}(t/\tau) e^{\frac{1}{2} i\gamma |A_0|^2 z}. \tag{3.2.46}$$

To get both sides to be equal, one requires

$$\frac{\beta_2}{\tau^2} + \gamma|A_0|^2 = 0 \implies |A_0|^2 = -\frac{\beta_2}{\gamma\tau^2}, \tag{3.2.47}$$

which requires $\beta_2 < 0$ as is consistent with the argument we made about SPM and dispersion imparting opposite chirp. With this constraint, we see that the left-hand side and right-hand side are equal to each other. We can understand the condition above as a constraint on the amplitude given known dispersion, pulse duration, and nonlinearity. Or we can understand it as a constraint on the parameters for a given amplitude.

Beyond the soliton solution described here, there is a continuous family of soliton solutions which are related by translations, phases, and initial velocities. I will not show it here, but you can show (with a bit more algebra) that the following is also a solution to the nonlinear Schrodinger equation:

$$A(z,t') = A_0 \text{sech}\left(\frac{t - t_0 + |\beta_2|\Omega_0 z}{\tau}\right) \exp\left[i\left(\phi_0 - \Omega_0(t-t_0) - \frac{1}{2}|\beta_2|\Omega_0^2 z + \frac{1}{2}\gamma|A_0|^2 z\right)\right]. \tag{3.2.48}$$

Compared to the soliton solution we evaluated previously, this one is translated by a time offset $t_0$ and has an overall phase $\phi_0$. Additionally there is a frequency offset $\Omega_0$ which gives the pulse an overall group velocity related to $|\beta_2|\Omega$ in the moving frame.

There also exist higher-order solitons which exhibit periodic breathing behavior that may be understood as collections of more than one soliton which nonlinearly interact.

## 3.3 Nonlinear refraction of beams

The treatment of nonlinear effects thus far has frozen out the transverse dynamics of the electromagnetic field: we have assumed that the light occupies effectively a single-mode waveguide. There are however very important effects that become apparent when we consider the transverse dynamics, including self-focusing, spatial solitons, and beam collapse. In what follows, we develop these effects. We will pursue a simplified treatment in which we consider propagation of scalar monochromatic beams. The topic of vectorial spatiotemporal pulses with transverse and longitudinal dynamics is an area of still active research.

The starting point is the Helmholtz equation for the positive frequency part of a monochromatic field at frequency $\omega$. We will consider the light to be propagating in a medium with uniform linear index $n_0$. Including nonlinear polarization, the result is

$$\left(\nabla^2 + k_0^2\right) E(\mathbf{r}) = -3\frac{\omega^2}{c^2}\chi^{(3)}|E(\mathbf{r})|^2 E(\mathbf{r}), \tag{3.3.1}$$

where we have defined $k_0 = n_0\omega/c$.

To proceed, let us consider a beam which propagates primarily along $z$ with wavevector $k_0$ with slow residual variations in $z$. While equivalent to our standard slowly varying envelope approximation, in the spatial case, this carries an additional implication. In particular, our slowly varying envelope approximation is equivalent to saying that there only negligible spatial Fourier components with large angles to the $z$ axis. This is called the *paraxial approximation*

because it says that the wavevector components of the beam are mostly parallel to the propagation axis . This is because, if we consider the Fourier transform of the field in space

$$E(\mathbf{k}) = \int d\mathbf{r}\ e^{i\mathbf{k}\cdot\mathbf{r}} E(\mathbf{r}), \tag{3.3.2}$$

the only $\mathbf{k}$ components we expect to see are ones satisfying

$$\mathbf{k}_\perp^2 + k_z^2 = k_0^2. \tag{3.3.3}$$

Therefore, if we insist that the $z$-dependence has Fourier components $k_z \approx k_0$, then we have that $|k_\perp| \ll k_0$, and so the angular spread of the beam must be small. This is because the wavevector components are associated with transverse propagation angles (in a small angle approximation)

$$\theta(k_\perp) \approx |k_\perp|/k_0 \ll 1, \tag{3.3.4}$$

establishing the equivalence of the spatially varying envelope approximation to the paraxial approximation.

Therefore we write

$$E(\mathbf{r}, \omega) = A(\boldsymbol{\rho}, z) e^{ik_0 z}, \tag{3.3.5}$$

where $A(\boldsymbol{\rho}, z)$ is a slowly varying envelope in $z$. Dealing with the $z$ derivatives as usual, we have that

$$\left(\nabla_\perp^2 + 2ik_0\partial_z\right) A(\boldsymbol{\rho}, z) = -3\frac{\omega^2}{c^2}\chi^{(3)}|A(\boldsymbol{\rho}, z)|^2 A(\boldsymbol{\rho}, z). \tag{3.3.6}$$

Rearranging terms, we find:

$$\partial_z A = \frac{i}{2k_0}\nabla_\perp^2 A + \frac{3i\omega\chi^{(3)}}{2n_0 c}|A|^2 A. \tag{3.3.7}$$

We then change units such that $|A|^2$ has dimensions of intensity rather than electric field squared (but we still refer to the variable as $A$). The resulting equation becomes

$$\partial_z A = \frac{i}{2k_0}\nabla_\perp^2 A + \frac{3i\omega\chi^{(3)}}{4\epsilon_0 n_0^2 c^2}|A|^2 A, \tag{3.3.8}$$

which we write as

$$\partial_z A = \frac{i}{2k_0}\nabla_\perp^2 A + i\kappa|A|^2 A, \tag{3.3.9}$$

where

$$\kappa = \frac{3\omega\chi^{(3)}}{4\epsilon_0 n_0^2 c^2} = \frac{n_2 k_0}{n_0}. \tag{3.3.10}$$

This equation will be the basis for our investigations of spatial nonlinear phenomena. Like the case where we studied nonlinear pulse propagation, we will start with the linear behavior. Then, we will consider nonlinearity without diffraction. And finally we will consider what happens when the two effects come together.

### 3.3.1 Diffraction of paraxial beams

In the absence of nonlinearity, we have

$$\partial_z A = \frac{i}{2k_0}\nabla_\perp^2 A. \tag{3.3.11}$$

To understand the dynamics of this equation, let us consider what happens when we have a known field $A(\boldsymbol{\rho}, 0)$ and we would like to know what the field is for $z > 0$. From Fourier considerations, we could write this as

$$A(\boldsymbol{\rho}, z) = \int \frac{d^2k}{(2\pi)^2} e^{i\mathbf{k}\cdot\boldsymbol{\rho} - i\frac{k^2}{2k_0}z} A(\mathbf{k}, 0), \tag{3.3.12}$$

where I have taken $\mathbf{k}_\perp \to \mathbf{k}$ for notational ease. A particularly important case is that of the *Gaussian beam*. This is described by a situation where

$$A(\boldsymbol{\rho}, 0) = A_0 e^{-\frac{\rho^2}{w_0^2}}, \tag{3.3.13}$$

where $w_0$ the *beam waist* is a measure of the width of the beam in the transverse directions. The resulting field at nonzero $z$ is given by the resulting Gaussian integral which is just a two-dimensionally separable form of the same integral we encountered when looking at the effect of second-order dispersion on pulses[9]. The result can be written as

$$A(\boldsymbol{\rho}, z) = A_0 \frac{1}{1 + iz/b} e^{-\frac{\rho^2}{w_0^2(1+iz/b)}}, \tag{3.3.14}$$

where $b = k_0 w_0^2/2$ is called the confocal parameter. The expression above for the so-called *Gaussian beam* is often expressed in a non-complex form as

$$A(\boldsymbol{\rho}, z) = A_0 \frac{1}{\sqrt{1 + z^2/b^2}} e^{-i\psi(z)} e^{-\frac{\rho^2}{w^2(z)}} e^{\frac{ik_0\rho^2}{2R(z)}}, \tag{3.3.15}$$

where

$$w(z) = w_0\sqrt{1 + \left(\frac{z}{b}\right)^2}, \tag{3.3.16}$$

$$R^{-1}(z) = \frac{z}{z^2 + b^2} \tag{3.3.17}$$

and

$$\psi(z) = \tan^{-1}\left(\frac{z}{b}\right). \tag{3.3.18}$$

This is the standard form of the Gaussian beam[10] Let us now discuss the physics of this solution. I should mention that this is one of the most important free-space propagating

[9] In general, the math of beams is quite analogous to that of pulses in the linear and nonlinear regimes even when one case has analytical solutions and the other does not

[10] If you compare this form to that in Haus' *Waves and Fields in Optoelectronics* you will find that ours maps to his with $\psi \to -\psi$ and $R \to -R$. This is a straightforward consequence of him using $e^{-ikz}$ to represent forward propagation while we use $e^{ikz}$. Additionally, you will find an additional factor of $\sqrt{\frac{2}{\pi w_0^2}}$ in his result because he defines a normalized Gaussian beam such that its square integral over $x, y$ is unity.

solutions to Maxwell equations as it very well approximates the fields emitted by laser cavities and thus many beams we work with are well-described by the Gaussian beam. Study it well!

The first thing to point out is that the beam width in the transverse direction, described by the beam waist $w(z)$, increases for $|z| > 0$, this is diffraction and is essentially equivalent to what occurs when we have light undergo single slit diffraction. The scale over which the beam waist changes is the confocal parameter $kw_0^2/2$. At large distance $z$ the waist expands linearly, implying that the light expands in a cone-like manner. The half-angle $\theta$ of that cone can be approximated as

$$\theta = \lim_{z\to\infty} \frac{w(z)}{z} = \frac{w_0}{b} = \frac{2}{k_0 w_0} = \frac{\lambda}{n_0 \pi w_0}. \tag{3.3.19}$$

This same form for the angle could be inferred from simple Fourier uncertainty principle considerations. Consider a wave in one dimension which is localized to a size $\Delta x \sim w_0$. Then there must also be a spread in wavevectors $\Delta k_x \sim \frac{1}{\Delta x} = \frac{1}{w_0}$, corresponding to an angular spread[11]. That angular spread is simply $\Delta\theta = \frac{\Delta k_x}{k_0} \sim \frac{\lambda}{n_0 w_0}$. Hence, we identify this Gaussian beam spreading as diffraction. Importantly, we see that the angle of expansion depends on the minimum beam waist. The more localized we are to start, the more the beam diverges as it propagates. This can be straightforwardly recalled using the Fourier uncertainty principle. We see that when $w_0 \gg \lambda$, the angular spread is low, and the paraxial approximation is satisfied. For beam waists close to the wavelength, the angular spread is large and this treatment becomes invalid. We need to consider vector diffraction theory in that case. Diffraction is expected by energy conservation to dilute the amplitude of the beam. This is exactly what the factor $(1 + z^2/b^2)^{-1/2}$ is doing.

The next factors we would like to understand are the phase factors. Let us start with the factor $e^{ik_0\rho^2/2R(z)}$. This describes a wavefront with a $z$-dependent radius of curvature $R(z)$. At large distances the surfaces of constant phase are given by parabolas with $z = \frac{k_0\rho^2}{2}$. The remaining factor $e^{-i\psi(z)}$ called the *Gouy phase* describes an additional effective wavevector for light at the beam waist (at large $z$ this factor is equivalent to an overall phase). That additional wavevector is of order $-1/b$ and essentially reflects that sharp transverse variations associated with localization must take away from the longitudinal wavevector.

Similar to our analysis of pulse propagation, we can define an instantaneous *spatial frequency* (also known as, wavevector) for the beam. Recall in the temporal case that we defined our instantaneous frequency in the context of finding the spectrogram of self-phase modulation. This instantaneous frequency arises from stationary phase considerations. The instantaneous wavevector is given by

$$\mathbf{k}(\mathbf{r}) = \nabla\left(k_0 z + \frac{ik_0\rho^2}{2R(z)} - \psi(z)\right) = \hat{z}\left(k_0 - \frac{b}{z^2+b^2} + \frac{k_0\rho^2}{2}\frac{z^2-b^2}{(z^2+b^2)^2}\right) + \frac{k_0}{R(z)}\boldsymbol{\rho}. \tag{3.3.20}$$

We will not worry about the two corrections to $k_0$ which are of the same order and are Gouy-like. Thus we will approximate the local wavevector as

$$\mathbf{k}(\mathbf{r}) \approx k_0\hat{z} + \frac{k_0}{R(z)}\boldsymbol{\rho}. \tag{3.3.21}$$

[11]The Fourier uncertainty principle is $\Delta x \Delta k_x \geq \frac{1}{2}$ which is straightforwardly inferred from taking the quantum mechanical version of the statement $\Delta x \Delta p_x \geq \frac{\hbar}{2}$ and taking $p_x = \hbar k_x$. I am dropping all prefactors since this argument is meant to just explain the scaling.

The local angle of propagation (if we think of our expanding beam in terms of a diverging bundle of rays) is, in a small-angle approximation:

$$\theta(\mathbf{r}) \approx \frac{\rho}{R(z)}, \tag{3.3.22}$$

which, at the beam waist, and for large $z$ approximates to $w(z)/z = w_0/b$ as consistent with our previous analysis. The instantaneous wavevector picture, besides giving a rigorous and intuitive ray-optics description of our beam, also will be indispensable for understanding the effect of nonlinearity, which we now turn to.

### 3.3.2 Self-phase modulation in space

Now that we understand the linear behavior somewhat, let us now consider the nonlinear terms alone. We are left with

$$\partial_z A = i\kappa |A|^2 A, \tag{3.3.23}$$

which can be seen to be solved by

$$A(\boldsymbol{\rho}, z) = A(\boldsymbol{\rho}, 0) \exp\left[i\kappa |A(\boldsymbol{\rho}, 0)|^2 z\right] A(\boldsymbol{\rho}, 0). \tag{3.3.24}$$

As we shall now show, this corresponds to changing the instantaneous wavevector of the beam, leading to a remarkable effect called self-focusing.

Let us consider what happens in the case of a Gaussian beam. Let us assume that the Gaussian beam is *collimated*: in other words, $k_0 w_0 \gg 1$ and so the confocal parameter is very large compared to the length of propagation. In this limit, we can approximate $z$ as zero for the Gaussian beam. In this case, this collimated beam experiences an additional phase (a phase modulation) of

$$\kappa |A_0|^2 e^{-2\rho^2/w_0^2}. \tag{3.3.25}$$

The instantaneous wavevector is therefore given by (for $z \ll b$)

$$\mathbf{k}(\mathbf{r}) = k_0 \hat{z} - 4\kappa z |A_0|^2 \frac{\boldsymbol{\rho}}{w^2} e^{-2\rho^2/w_0^2}. \tag{3.3.26}$$

You can already see it: the additional wavevector points inwards towards the center of the beam at all points along the beam. That means that the light rays want to bend inwards towards the center, making the beam smaller. This is the essence of self-focusing. If it is possible for example for all rays to converge onto a point, then the beam will undergo a catastrophic collapse. In particular: the beam having some intensity causes it to bend inwards, and increase its intensity. That however causes more bending, until the beam focuses to a point. At this stage, the beam intensity can become so high that few materials can withstand it and undergo damage.

Before we provide a condition for self-focusing, it is worthwhile to describe another perspective on how we see inward focusing that does not rely on the instantaneous wavevector. The intensity-dependent phase modulation is strongest at the center corresponding to a phase advance for rays propagating on-axis relative to rays propagating (along $z$) which are displaced from the beam axis. If we draw surfaces of constant phase, they will be curved, and

the curvature will be such that they appear to emanate from a focus at positive $z$. This is suggestive that the nonlinearity is acting as a focusing lens.

In what follows, we will pursue a significantly oversimplified analysis of self-focusing which should not be taken to be quantitative. However, it will yield the correct scaling laws for the so-called critical power for self-focusing, and the length over which self-focusing occurs. We see that the inward bending of rays depends on the phase gradient. Let us approximate this phase gradient as $\kappa I z/w$ where $I$ is some characteristic intensity and $w$ is some characteristic length. For a Gaussian beam, $I$ could be the intensity at the beam center and $w$ the minimum beam waist. In this case, the downward bending angle of an off-axis ray is given by

$$\theta_{\mathrm{NL}} = \frac{\kappa I z}{w k_0} = \frac{n_2 I z}{n_0 w}. \tag{3.3.27}$$

We shall (very coarsely) assume that this angle is maintained for a distance $z$ until the ray crosses the propagation axis, therefore traversing a lateral distance $w$. By construction, this ray always meets the central on-axis ray since from our considerations above for the Gaussian beam, the central ray experiences no deflection. Then $\theta \approx w/z$ in a small-angle approximation and we have

$$\theta_{\mathrm{NL}}^2 = \frac{n_2 I}{n_0} \implies \theta_{\mathrm{NL}} = \sqrt{\frac{n_2 I}{n_0}}. \tag{3.3.28}$$

The distance required to focus is given by

$$L_f = w/\theta_{\mathrm{NL}} = w\sqrt{\frac{n_0}{n_2 I}}. \tag{3.3.29}$$

Now one thing our analysis has neglected so far is the fact that a Gaussian beam will naturally spread outwards with some asymptotic angle $\theta_{\mathrm{D}} = \frac{\lambda}{\pi n_0 w_0}$. One can expect that if $\theta_{\mathrm{NL}} < \theta_{\mathrm{D}}$ then the beam will still spread out, just at a reduced angle. Meanwhile, if $\theta_{\mathrm{NL}} > \theta_{\mathrm{D}}$, then one gets catastrophic self-focusing. The boundary case is when self-focusing balances diffraction, and in this case it is possible to get a shape-preserving beam called an *spatial soliton*. This effect is called self-trapping and this occurs for a critical intensity $I_c$ and corresponding critical power $P_c = \frac{\pi}{2} w^2 I$ equal to[12]:

$$\sqrt{\frac{n_2 I_c}{n_0}} = \frac{\lambda}{\pi n_0 w} \implies P_c = \frac{\lambda^2}{2\pi n_0 n_2}. \tag{3.3.30}$$

Expressed in terms of the critical power, we can write the self-focusing length as[13]

$$L_f = \frac{\pi n_0 w^2}{\lambda} \frac{1}{\sqrt{P/P_c}}. \tag{3.3.31}$$

For $P = P_c$ and a 100 micron beam waist at a wavelength of 1 micron with a characteristic index of 1.5, the self-focusing length is roughly 5 cm. The corresponding critical power in a material such as silica is on the order of 10 MW.

[12]The prefactor for this critical power is different than both what is in Boyd and what is in Yariv's *Quantum Electronics*. This is to say that we should only take the scaling seriously.

[13]This also slightly differs from Boyd in overall prefactor, as is to be expected.

### 3.3.3 Filamentation of an optical beam

We conclude this chapter by discussing what happens when the beam power is significantly larger than the critical power for self-focusing. The short answer is that the beam will break up into beamlets or filaments each which carry approximately the critical power for self-trapping. We will pursue a simplified analysis of this problem as follows. Let us crudely approximate our beam as a strong plane wave. The rough idea is that the beam will break up into other beams propagating in other directions. So let us describe the growth of these beamlets as follows. Let us take our field as

$$A(\boldsymbol{\rho}, z) = \left(A_0(z) + \delta A(\boldsymbol{\rho}, z)\right), \tag{3.3.32}$$

where $|\delta A| \ll |A_0|$. In other words we have a strong forward-propagating plane wave representing the main beam[14] which seeds a transverse varying field $\delta A(\boldsymbol{\rho}, z)$.

$$\partial_z A_0 = i\kappa |A_0|^2 A_0 \implies A_0(z) = A_0(0) e^{i\kappa |A_0(0)|^2 z} \equiv A_0 e^{iQz}. \tag{3.3.33}$$

In what follows, we will refer to $A_0(0)$ as $A_0$ and $Q = \kappa |A_0|^2$. At order $\delta A$, we have two coupled equations

$$\begin{aligned} \partial_z \delta A &= \frac{i}{2k_0} \nabla_\perp^2 \delta A + i\kappa \left(2|A_0(z)|^2 \delta A + A_0^2(z) \delta A^*\right) \\ \partial_z \delta A^* &= -\frac{i}{2k_0} \nabla_\perp^2 \delta A^* - i\kappa \left(2|A_0(z)|^2 \delta A^* + A_0^{*2}(z) \delta A\right). \end{aligned} \tag{3.3.34}$$

which may be simplified as

$$\begin{aligned} \partial_z \delta A &= \frac{i}{2k_0} \nabla_\perp^2 \delta A + i\kappa \left(2|A_0|^2 \delta A + A_0^2 e^{2iQz} \delta A^*\right) \\ \partial_z \delta A^* &= -\frac{i}{2k_0} \nabla_\perp^2 \delta A^* - i\kappa \left(2|A_0|^2 \delta A^* + A_0^{*2} e^{-2iQz} \delta A\right). \end{aligned} \tag{3.3.35}$$

By defining envelope variables $\delta A = \delta \tilde{A} e^{iQz + i\arg(A_0)}$, we can get rid of the nonlinear oscillations and write

$$\begin{aligned} \partial_z \delta \tilde{A} &= \frac{i}{2k_0} \nabla_\perp^2 \delta \tilde{A} + i\kappa \left(|A_0|^2 \delta \tilde{A} + |A_0|^2 \delta \tilde{A}^*\right) \\ \partial_z \delta \tilde{A}^* &= -\frac{i}{2k_0} \nabla_\perp^2 \delta \tilde{A}^* - i\kappa \left(|A_0|^2 \delta \tilde{A}^* + |A_0|^2 \delta \tilde{A}\right). \end{aligned} \tag{3.3.36}$$

Let us now go into the Fourier domain in the transverse wavevector space, defining $A(\boldsymbol{\rho}, z) = \int \frac{d^2q}{(2\pi)^2} e^{i\mathbf{q}\cdot\boldsymbol{\rho}} A(\mathbf{q}, z)$. We see then that we may write

$$\partial_z \begin{pmatrix} \tilde{A}(\mathbf{q}, z) \\ \tilde{A}(-\mathbf{q}, z)^* \end{pmatrix} = i \begin{pmatrix} K & Q \\ -Q & -K \end{pmatrix} \begin{pmatrix} \tilde{A}(\mathbf{q}, z) \\ \tilde{A}(-\mathbf{q}, z)^* \end{pmatrix}. \tag{3.3.37}$$

In writing this, we have used the fact that $(A^*)(\mathbf{q}) = (A(-\mathbf{q}))^*$. We have also defined $K = Q - \frac{q^2}{2k_0}$.

[14] Remember that the overall electric field has an $e^{ik_0 z}$.

As with typical linear systems of equations, a good way to analyze it is by looking at the eigenvalues and eigenvectors of the matrix. The eigenvalues $\lambda_{1,2}$ are found to be[15]

$$\lambda_{1,2} = \pm\sqrt{Q^2 - K^2} = \pm\sqrt{2\frac{q^2}{2k_0}Q - \left(\frac{q^2}{2k_0}\right)^2} = \sqrt{\frac{q^2}{2k_0}}\sqrt{2Q - \frac{q^2}{2k_0}}. \tag{3.3.38}$$

We see that in the absence of nonlinearity, the eigenvalues are purely imaginary, corresponding to oscillatory solutions. For finite nonlinearity, we see that for small transverse wavevectors, there is a positive eigenvalue corresponding to an exponentially growing solution. This corresponds to the exponential growth of perturbations from initial noise. The maximum growth rate occurs for a transverse wavevector $q^2 \equiv 2k_0 x$ which maximizes

$$x(2Q - x), \tag{3.3.39}$$

for which the maximum is simply $x = Q$, corresponding to a maximum gain at wavevector $q_{\max}$ equal to

$$q_{\max} = \sqrt{2k_0\kappa|A_0|^2}, \tag{3.3.40}$$

corresponding to the harmonic mean of the central wavevector and the "nonlinear wavevector".

The picture we have developed is now this: a plane wave propagating in a nonlinear medium is unstable and seeds the exponential growth of waves propagating off-axis. We can go back to the beam picture by considering each of these plane waves as beamlets carrying a localized region of light around it[16]. These beamlets in reality will diffract while growing. This puts a natural cap on the growth because at some point the power in each beamlet will approach the critical power for self-trapping. Thus the initial beam develops modulations which grow into beams or filaments which carry power approximately equal to $P_c$. This is called filamentation, and the growth of transverse variations (modulations) from noise is called *modulation instability*, because if there is any initial modulation present in the field (nonzero $\delta A$ in our analysis), then the perturbations will grow from noise.

We mentioned that the perturbations grow from noise. This can correspond to imperfections such as aberrations in the optical beam, or propagation through even a weakly disordered medium. Even in an ostensibly clean beam with field $A_0$ only, with no initial perturbation, it turns out these perturbations can continue to grow. This happens due to *quantum noise*: Heisenberg's uncertainty principle for the electric and magnetic field states that the fields cannot be exactly zero, only on average both zero. So we can think there is a small fluctuating perturbation called quantum noise which seeds this filamentation. That brings us to the second part of the course, on the *quantum* description of nonlinear optical phenomena.

[15]The quick way to do this is to note that the matrix is $iK\sigma_z - Q\sigma_y$ and to use the standard result for eigenvalues of sums of Pauli matrices. Note however that because of the imaginary coefficient of $\sigma_z$, the matrix is not Hermitian.

[16]The linearized analysis above is much more complicated with an optical beam although it can still be done

# Part 2: Quantum Nonlinear Optics

# Chapter 4

# Quantum mechanical description of the electromagnetic field in matter

We have thus far spent our time in this course studying how the propagation of light is modified by a material. In the linear regime, in which the material polarization is linear in the total electric field, the overall effect is to establish a refractive index for light. In the nonlinear regime in which the material polarization is nonlinear, a variety of new effects emerge such as the generation of new frequencies, the electro-optic effect, and the intensity-dependent refractive index. In all of our treatment thus far, our treatment of the electromagnetic field and matter has been according to classical physics, although we have explained a few things in terms of a heuristic photon picture.

At the same time, the fundamental description of light and its interaction with matter is based on quantum mechanics. In standard treatments of quantum mechanics, it is typical to quantize matter: to quantize electrons, nuclei, and other matter particles and study their quantum properties based on the dynamics of the corresponding wavefunction. Based on the time-development of the wavefunction, expectation values of Hermitian operators (observables) can be computed. It is in some sense only expectation values of Hermitian operators that we can physically observe. Although it is not typical in standard courses of quantum mechanics to quantize the light field, it can be done and conceptually the quantum mechanics of the electromagnetic field is similar to the quantum mechanics of matter. Compared to electromagnetic field quantization as is done for example in quantum field theory, our quantization has to be able to describe the quantum properties of light propagating in matter, which requires accounting for the effect of polarization in a quantum mechanically consistent way.

In this second part of the course (Units 4 and 5), we will treat the electromagnetic field quantum mechanically. Such a treatment is necessary to describe the statistical properties of light, which are important for describing spontaneous emission, noise in light sources, fundamental limitations to optical amplifiers, and nonclassical correlations such as entanglement and squeezing which are of importance for quantum-enhanced sensing.

In Unit 4, we start by quantizing the electromagnetic field in matter. The outcome of this procedure is a Hamiltonian for the electromagnetic field expressed in terms of electromagnetic field *operators*: specifically the vector potential and the displacement field, which will have canonical commutation relations much like the position and momentum of a particle. Using this

Hamiltonian, we will explore the stationary states (eigenstates) and important superpositions of them which correspond to commonly realized quantum states of light. From there, we will describe how we measure the quantum properties of light, and in doing so, develop a theory of photodetection. With the apparatus of photodetection theory, we can then describe different types of correlation measurements of light and under what conditions a certain measurement cannot be realized classically [1].

In many cases, light states experience attenuation or amplification which correspond to *non-conservative* or *open system* dynamics classically. A quantum mechanically consistent treatment of such open system dynamics requires the introduction of *quantum noise sources* whose effect is typically to degrade quantum correlations and degrade signal-to-noise ratios. Such effects lead to important limits on devices such as optical amplifiers, with important implications for optical communications.

In Unit 5, on the quantum optical description of nonlinear optical phenomena, we will use the apparatus developed in this unit describe how nonlinear media lead to the generation of quantum mechanical states of light such as entangled photons and squeezed light. And we will conclude by illustrating how quantum light states allow measurement sensitivities which can exceed what is possible classically.

## 4.1 Quantization of the electromagnetic field in matter

### 4.1.1 Electromagnetic Lagrangian of a lossless nonlinear medium

We start by developing a quantum description of the electromagnetic field in a material. Such a description is called *macroscopic quantum electrodynamics*. We follow a standard procedure in quantum field theory: (a) identify a Lagrangian whose Euler-Lagrange equations reproduce the classical equations of motion, (b) identify the corresponding Hamiltonian via the Legendre transform, and (c) canonically quantize the fields by introducing canonical commutation relations for the complementary dynamical variables. These canonical commutation relations must reproduce the classical description in the appropriate classical limit. This procedure is called canonical quantization. We will assume familiarity with Lagrangian and Hamiltonian mechanics, as well as Maxwell's equations based on the vector and scalar potentials. These main results are reviewed in Appendices 1 and 2, and the reader is encouraged to look there for more details.

In Unit 1, we discussed the energy of the electromagnetic field in a nonlinear medium and in so doing, introduced a Lagrangian for the electromagnetic field in matter which reproduced the Maxwell equations in the presence of polarization. We found that the Lagrangian density was given in terms of the electric and magnetic fields via

$$\mathcal{L} = \frac{\epsilon_0}{2}(\mathbf{E}^2 - c^2\mathbf{B})^2 + \mathcal{L}_{\text{pol}}, \tag{4.1.1}$$

where

$$\mathcal{L}_{\text{pol}} = \epsilon_0 \sum_{n=1}^{\infty} \frac{1}{n+1} \chi^{(n)}_{i_0 i_1 i_2 \cdots i_n} E_{i_0} E_{i_1} \cdots E_{i_n} \equiv \epsilon_0 \sum_{n=1}^{\infty} \frac{1}{n+1} \chi^{(n)} : \mathbf{E}^{\otimes(n+1)}. \tag{4.1.2}$$

[1] This chapter was written with helpful contributions from Jamison Sloan.

Let us now find the corresponding Euler-Lagrange equations for this Lagrangian. Before we do this, we will do a change of variables, from $\mathbf{E}$ and $\mathbf{B}$ to $\mathbf{A}$ and $\phi$, the vector and scalar potentials. As a reminder, the fields are related to the potentials by $\mathbf{E} = -\partial_t \mathbf{A} - \nabla\phi$ and $\mathbf{B} = \nabla \times \mathbf{A}$. Importantly, we remember from electrodynamics that there is *gauge freedom*: we are allowed to change the potentials by a gauge transformation without changing the physical electric and magnetic fields. Recall that if we take $\phi \to \phi + \partial_t \chi$ and $\mathbf{A} \to \mathbf{A} - \nabla\chi$ for any function $\chi$, we get the same fields. Therefore, when we work with potentials, it is useful to specify a gauge. A useful gauge is the so-called *Coulomb gauge* in which $\nabla \cdot \mathbf{A} = 0$[2].

Quantization is usually approached from the standpoint of the potentials because there are certain constraints which emerge naturally in the language of potentials that are important to track. We will see this shortly. The Euler-Lagrange equations for the scalar potential are[3]:

$$\frac{\partial \mathcal{L}}{\partial \phi} + \partial_t \frac{\partial \mathcal{L}}{\partial(\partial_t \phi)} + \partial_j \frac{\partial \mathcal{L}}{\partial(\partial_j \phi)} = 0, \tag{4.1.3}$$

where the $j$ denotes spatial indices which are summed over. The first and second terms are zero, as the only way $\phi$ appears in the equation is through its gradient. The relevant derivative is most easily evaluated by

$$\frac{\partial \mathcal{L}}{\partial(\partial_j \phi)} = \frac{\partial \mathcal{L}}{\partial E_i}\frac{\partial E_i}{\partial(\partial_j \phi)} = -\delta_{ij}\frac{\partial \mathcal{L}}{\partial E_i}. \tag{4.1.4}$$

The derivative with respect to the electric field is evaluated as

$$\frac{\partial \mathcal{L}}{\partial E_i} = \left(\epsilon_0 E_i + \epsilon_0 \sum_{n=1}^{\infty} \chi^{(n)}_{ii_1\cdots i_n} E_{i_1} \cdots E_{i_n}\right) = D_i, \tag{4.1.5}$$

where $\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}$. In evaluating the derivative, I made use of the Kleinman symmetry of a lossless medium which allows us to freely permute the indices of $\chi^{(n)}$. This renders all $n+1$ terms generated in the product rule equal to each other. Combining these derivatives with the Euler-Lagrange equation gives

$$\partial_j \frac{\partial \mathcal{L}}{\partial(\partial_j \phi)} = -\epsilon_0 \partial_j D_j = 0, \tag{4.1.6}$$

In other words, we get Gauss' law:

$$\nabla \cdot \mathbf{D} = 0. \tag{4.1.7}$$

This equation tells us that $\phi$ is not freely determined. Why? Because we can write

$$\nabla \cdot \mathbf{D} = \nabla \cdot (\epsilon_0(-\partial_t \mathbf{A} - \nabla\phi) + \mathbf{P}). \tag{4.1.8}$$

In the Coulomb gauge, we may write

$$\nabla \cdot \mathbf{D} = -\epsilon_0 \nabla^2 \phi - \rho_b = 0 \implies \nabla^2 \phi = -\frac{\rho_b}{\epsilon_0}. \tag{4.1.9}$$

[2]The reader should convince themselves that if they are in a gauge in which $\nabla \cdot \mathbf{A} \neq 0$ then they can find a $\chi$ which makes the vector potential divergenceless and that this $\chi$ satisfies a Poisson equation.

[3]As described in the relevant Appendix, we treat the field, its time derivatives, and its space derivatives all as independent variables.

where $\rho_b = \nabla \cdot \mathbf{P}$ is the bound polarization charge. Therefore the potential is given in terms of the electric field itself (which determines uniquely the polarization) by Coulomb's law (this is why the gauge is called Coulomb gauge).

So much for the scalar potential. Let us now look at the equation for the vector potential. For each component, $A_i$, the Euler-Lagrange equation is

$$\frac{\partial \mathcal{L}}{\partial A_i} + \partial_t \frac{\partial \mathcal{L}}{\partial(\partial_t A_i)} + \partial_j \frac{\partial \mathcal{L}}{\partial(\partial_j A_i)} = 0. \tag{4.1.10}$$

The first term vanishes while the other two are non-zero. The first of the two is

$$\frac{\partial \mathcal{L}}{\partial(\partial_t A_i)} = \frac{\partial \mathcal{L}}{\partial E_j}\frac{\partial E_j}{\partial(\partial_t A_i)} = -\delta_{ij}\frac{\partial \mathcal{L}}{\partial E_j} = -D_i. \tag{4.1.11}$$

The second of the two is evaluated using a similar chain-rule approach as:

$$\frac{\partial \mathcal{L}}{\partial(\partial_j A_i)} = \frac{\partial \mathcal{L}}{\partial B_k}\frac{\partial B_k}{\partial_j A_i} = -\frac{B_k}{\mu_0}\frac{\partial B_k}{\partial_j A_i}. \tag{4.1.12}$$

The derivative of the magnetic field is evaluated as

$$\frac{\partial B_k}{\partial_j A_i} = \frac{\partial}{\partial_j A_i}\epsilon_{krs}\partial_r A_s = \epsilon_{krs}\delta_{jr}\delta_{is} = \epsilon_{kji}, \tag{4.1.13}$$

where $\epsilon_{ijk}$ is the usual Levi-Civita symbol. Using this, we find

$$\frac{\partial \mathcal{L}}{\partial(\partial_j A_i)} = -\frac{B_k}{\mu_0}\epsilon_{kji}, \tag{4.1.14}$$

and so the Euler-Lagrange equation for the vector potential may be written as

$$-\partial_t D_i - \frac{1}{\mu_0}\epsilon_{kji}\partial_j B_k = 0 \implies \nabla \times \mathbf{H} = \partial_t \mathbf{D}, \tag{4.1.15}$$

where $\mathbf{H} = \mathbf{B}/\mu_0$. This is just the Ampere-Maxwell law in a medium. We may use these results to get an overall wave equation as follows. From the Ampere-Maxwell law and the definition of the potentials, we have

$$\nabla \times \nabla \times \mathbf{A} = \mu_0 \partial_t(\epsilon_0 \mathbf{E} + \mathbf{P}) \implies -\nabla \times \nabla \times \partial_t \mathbf{A} = -\frac{1}{c^2}\partial_t^2 \mathbf{E} - \mu_0 \partial_t^2 \mathbf{P}. \tag{4.1.16}$$

Using $-\partial_t \mathbf{A} = \mathbf{E} + \nabla\phi$ and $\nabla \times \nabla\phi = 0$, we have

$$\nabla \times \nabla \times \mathbf{E} + \frac{1}{c^2}\partial_t^2 \mathbf{E} = -\mu_0 \partial_t^2 \mathbf{P}, \tag{4.1.17}$$

which is the Maxwell wave equation. From these, we can in principle back-track and get the magnetic Gauss law and the Faraday laws as well.

We have now shown that our postulated Lagrangian reproduces the classical Maxwell equations. Before quantizing the equations, we now develop the expression for the Hamiltonian, which is the classical energy of the system. You also know however that the Hamiltonian is the generator of time-evolution in quantum mechanics, and so the Hamiltonian will be a central object for us in our quantization.

### 4.1.2 Hamiltonian of a lossless nonlinear medium

The Hamiltonian $H$ is typically expressed as an integral over the Hamiltonian density $\mathcal{H}$ (energy density) via

$$H = \int d^3r \; \mathcal{H}. \tag{4.1.18}$$

The Hamiltonian density follows from a Legendre transform of the Lagrangian. You may recall for a single particle, $H(p,q) = p\dot{q} - L(q,\dot{q})$ where $p = \frac{\partial L}{\partial \dot{q}}$ is the canonical momentum corresponding to the generalized coordinate $q$. The idea in field theory is the same where in our case the generalized coordinates are taken to be the potentials $\phi, \mathbf{A}$. The Hamiltonian density corresponding to our electromagnetic Lagrangian is:

$$\mathcal{H} = \frac{\partial \mathcal{L}}{\partial(\partial_t \phi)}(\partial_t \phi) + \frac{\partial \mathcal{L}}{\partial(\partial_t A_i)}(\partial_t A_i) - \mathcal{L}. \tag{4.1.19}$$

The canonical momentum for the scalar potential

$$\Pi_0 = \frac{\partial \mathcal{L}}{\partial(\partial_t \phi)} = 0, \tag{4.1.20}$$

which is a manifestation of the fact that $\phi$ is not an independent degree of freedom. The canonical momentum density for the vector potential however is nontrivial and given by

$$\Pi_i = \frac{\partial \mathcal{L}}{\partial(\partial_t A_i)} = -D_i. \tag{4.1.21}$$

Very importantly, the canonical momentum is the displacement field. The Hamiltonian density may now be written as

$$\mathcal{H} = -D_i(-E_i - \partial_i \phi) - \mathcal{L} = \mathcal{H}' + \mathbf{D} \cdot \nabla \phi, \tag{4.1.22}$$

where $\mathcal{H}' = \mathbf{D} \cdot \mathbf{E} - \mathcal{L}$. I have roped off the last term off because it does not contribute to the Hamiltonian. That is because

$$\int d^3r \; \mathbf{D} \cdot \nabla \phi = -\int d^3r \; \phi(\nabla \cdot \mathbf{D}) = 0. \tag{4.1.23}$$

Therefore, we will not keep track of it, and only consider

$$\mathcal{H}' = \mathbf{D} \cdot \mathbf{E} - \mathcal{L} = \epsilon_0 \sum_{n=1}^{\infty} \frac{n}{n+1} \epsilon^{(n)} : \mathbf{E}^{\otimes(n+1)} + \frac{\mathbf{B}^2}{2\mu_0}, \tag{4.1.24}$$

where

$$\epsilon^{(n)} = \chi^{(n)} \text{ if } n \geq 2; \text{ else } \epsilon^{(1)} = 1 + \chi^{(1)}, \tag{4.1.25}$$

where "1" represents the two-by-two identity matrix.

To quantize the fields, we follow the standard procedure in quantum mechanics, which is to promote the canonical positions and momenta to operators satisfying canonical commutation relations. To do this, we start by expressing the Hamiltonian directly in terms of the

canonically conjugate variables which are $A_i, \Pi_i = -D_i$. This is done as follows: let us define the relationship $\mathbf{E}(\mathbf{D})$ via

$$\mathbf{E} = \sum_n \eta^{(n)} : \mathbf{D}^{\otimes n}. \tag{4.1.26}$$

The values of $\eta$ are expressed in terms of the nonlinear susceptibilities $\chi$ by writing $\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}$ and solving the resulting equation order by order in $\mathbf{E}$ (see footnote for example[4].)

To express the Hamiltonian density $\mathcal{H}'$ in terms of $\mathbf{D}$, we make use of the following identity[5]:

$$\int \mathbf{E} \cdot d\mathbf{D} = \frac{1}{2}\epsilon_0 \mathbf{E}^2 + \epsilon_0 \sum_{n=1}^{\infty} \frac{n}{n+1} \chi^{(n)} : \mathbf{E}^{\otimes(n+1)} = \epsilon_0 \sum_{n=1}^{\infty} \frac{n}{n+1} \epsilon^{(n)} : \mathbf{E}^{\otimes(n+1)}. \tag{4.1.29}$$

Using this result, and expressing $\mathbf{E}$ in terms of $\mathbf{D}$ and evaluating the resulting integral (via the same manipulations as in Footnote 4), one arrives at

$$H = \int d^3 r \ \sum_{n=1}^{\infty} \eta^{(n)} : \mathbf{D}^{\otimes(n+1)} + \frac{(\nabla \times \mathbf{A})^2}{2\mu_0}, \tag{4.1.30}$$

where I have expressed $\mathbf{B}$ in terms of $\mathbf{A}$ to express the Hamiltonian fully in terms of $\mathbf{D}, \mathbf{A}$.

### 4.1.3 Quantization

The canonical quantization proceeds by promoting $\mathbf{A}$ and $\mathbf{D}$ to operators satisfying canonical commutation relations. Since we can think of fields as a collection of $c$ numbers for each position and each direction, it is tempting to impose the commutation relation $[A_i(\mathbf{r}), \Pi_j(\mathbf{r}')] = [A_i(\mathbf{r}), -D_j(\mathbf{r}')] = i\hbar\delta_{ij}\delta(\mathbf{r} - \mathbf{r}')$. However this cannot be right as it is not consistent with $\nabla \cdot \mathbf{A} = 0$ and $\nabla \cdot \mathbf{D} = 0$. In fact, since both fields are divergenceless, the resulting tensor function needs to project functions onto other divergenceless (transverse) functions. The corresponding answer is instead

$$[D_i(\mathbf{r}), A_j(\mathbf{r}')] = i\hbar\delta_{ij}^{\perp}(\mathbf{r} - \mathbf{r}'). \tag{4.1.31}$$

The object $\delta_{ij}^{\perp}(\mathbf{r} - \mathbf{r}')$ is called the *transverse delta function* and is a distribution defined by

$$\int d^3 r' \ \delta_{ij}^{\perp}(\mathbf{r} - \mathbf{r}') X_j(\mathbf{r}') = X_i^{\perp}(\mathbf{r}), \tag{4.1.32}$$

[4]The results for the first two terms are, for example,

$$\eta_{ij}^{(1)} = \frac{1}{\epsilon_0}(1 + \chi^{(1)})_{ij}^{-1} \tag{4.1.27}$$

$$\eta_{ijk}^{(2)} = -\epsilon_0 \eta_{il}^{(1)} \chi_{lmn}^{(2)} \eta_{mj}^{(1)} \eta_{nk}^{(1)}. \tag{4.1.28}$$

We see, for example, that the second order response $\eta^{(2)}$ is proportional to $\chi^{(2)}$, but now contains some extra factors related to the linear response. The third-order response $\eta^{(3)}$ will similarly involve $\eta^{(1)}$ and $\eta^{(2)}$

[5]This identity is proved as follows: write $\int \mathbf{E} \cdot d\mathbf{D} = \epsilon_0 \int \mathbf{E} \cdot d\mathbf{E} + \int \mathbf{E} \cdot d\mathbf{P}$. The first term is $\frac{1}{2}\epsilon_0 \mathbf{E}^2$. The second term is integrated by noting that $d\mathbf{P} = \sum_n d\mathbf{P}^{(n)} = n\mathbf{P}^{(n)} \cdot d\mathbf{E}$. The factor of $n$ follows from Kleinman symmetry. The remaining integral may be done by similarly noting that $\mathbf{P}^{(n)} \dot{d}\mathbf{E} = \frac{1}{n+1} d(\chi^{(n)} : \mathbf{E}^{\otimes(n+1)})$.

where $\mathbf{X}^\perp(\mathbf{r})$ is the divergence-less (transverse) part of $\mathbf{X}$ defined such that $\nabla \cdot \mathbf{X}^\perp = 0$. Recall from electrodynamics that the Helmholtz-Hodge decomposition enables us to decompose an arbitrary differentiable vector function into a divergence-less (transverse) part and a curl-less (longitudinal) part[6]. We will take this commutator as the fundamental postulate of macroscopic quantum electrodynamics. With this, and the Hamiltonian, all else follows.

It will turn out that in many cases, we want commutators of the displacement field and the magnetic field. That commutator is given by:

$$[D_i(\mathbf{r}), B_j(\mathbf{r}')] = i\hbar\epsilon_{jlm}\partial_l^{'}\delta_{im}^{\perp}(\mathbf{r}-\mathbf{r}'), \tag{4.1.34}$$

where $\partial'$ denotes derivative with respect to $\mathbf{r}'$.

### 4.1.4 Heisenberg picture description of the quantized electromagnetic field

To check that this commutator and Hamiltonian are valid, we now derive the Heisenberg equations of motion and show that in the classical limit, they reduce exactly to Maxwell's equations in a nonlinear medium. Recall that the Heisenberg equations are generally expressed as

$$\dot{O} = \frac{i}{\hbar}[H, O], \tag{4.1.35}$$

where $O$ is some operator. Let us start by examining the time-development of $\mathbf{D}$, which satisfies

$$\partial_t D_j(\mathbf{x}) = \frac{i}{\hbar}[H, D_j(\mathbf{x})] = \frac{i}{2\hbar\mu_0}\int d^3r\ [B_k(\mathbf{r})^2, D_j(\mathbf{x})], \tag{4.1.36}$$

which using the fundamental commutator can be written as

$$\partial_t D_j(\mathbf{x}) = \frac{i}{\hbar\mu_0}\int d^3r\ B_k(\mathbf{r})(-i\hbar\epsilon_{klm}\partial_l\delta_{jm}^{\perp}(\mathbf{r}-\mathbf{x})), \tag{4.1.37}$$

which can be written as

$$\partial_t D_j(\mathbf{x}) = \frac{1}{\mu_0}\int d^3r\ (\nabla\times\mathbf{B})_m\delta_{jm}^{\perp}(\mathbf{r}-\mathbf{x})) = \frac{1}{\mu_0}(\nabla\times\mathbf{B})_j(\mathbf{x}). \tag{4.1.38}$$

In other words, we have

$$\nabla\times\mathbf{H} = \partial_t\mathbf{D}, \tag{4.1.39}$$

which is the Ampere-Maxwell law in operator form.

Let us now derive an equation for the time-development of the magnetic field. That equation is

$$\partial_t B_j(\mathbf{x}) = \frac{i}{\hbar}[H, B_j(\mathbf{x})] = \frac{i}{\hbar}\int d^3r\ \sum_{n=1}^{\infty}\frac{1}{n+1}[\eta^{(n)} : \mathbf{D}(\mathbf{r})^{\otimes(n+1)}, B_j(\mathbf{x})]. \tag{4.1.40}$$

[6]The names transverse and longitudinal are most easily understood by noting that a divergence-less function, in Fourier space, satisfies $\mathbf{k}\cdot\mathbf{X}(\mathbf{k}) = 0$ while a curl-less function satisfies $\mathbf{k}\times\mathbf{X}(\mathbf{k}) = 0$. Therefore a transverse (longitudinal) function, in Fourier space, has the property that the vector field $\mathbf{X}(\mathbf{k})$ is transverse (longitudinal) to $\mathbf{k}$. From this reasoning, you can convince yourself that in Fourier space the transverse delta function is represented by

$$\delta_{ij}^{\perp}(\mathbf{r}-\mathbf{r}') = \int\frac{d^3k}{(2\pi)^3}e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}(\delta_{ij}-\hat{k}_i\hat{k}_j). \tag{4.1.33}$$

The tensor structure precisely projects out the part of a vector transverse to $\mathbf{k}$.

Using the product rule for commutators (e.g., $[AB, C] = A[B, C] + [A, C]B$) repeatedly, the above reduces to

$$\partial_t B_j(\mathbf{x}) = \frac{i}{\hbar} \int d^3r \ \sum_{n=1}^{\infty} (\eta^{(n)} : \mathbf{D}(\mathbf{r})^{\otimes n})_{i_0} \times i\hbar \epsilon_{jlm} \partial_l^{'} \delta^{\perp}_{i_0 m}(\mathbf{r} - \mathbf{x}), \tag{4.1.41}$$

where $(\eta^{(n)} : \mathbf{D}(\mathbf{r})^{\otimes n})_{i_0} = \eta^{(n)}_{i_0 i_1 \cdots i_n} D_{i_1} \cdots D_{i_n}$ and the primed derivative denotes differentiation with respect to $\mathbf{x}$. To simplify this further, we note that since $E_{i_0} = \sum_{n=1}^{\infty} (\eta^{(n)} : \mathbf{D}(\mathbf{r})^{\otimes n})_{i_0}$, we my write

$$\partial_t B_j(\mathbf{x}) = \frac{i}{\hbar} \int d^3r \ E_{i_0}(\mathbf{r}) \times i\hbar \epsilon_{jlm} \partial_l^{'} \delta^{\perp}_{i_0 m}(\mathbf{r} - \mathbf{x}). \tag{4.1.42}$$

To proceed, we write

$$\partial_t B_j(\mathbf{x}) = -\epsilon_{jlm} \partial_l^{'} \int d^3r \ E_{i_0}(\mathbf{r}) \times \delta^{\perp}_{i_0 m}(\mathbf{r} - \mathbf{x}) = -\epsilon_{jlm} \partial_l^{'} E^{\perp}_m(\mathbf{x}) = -(\nabla \times \mathbf{E}^{\perp})_j(\mathbf{x}). \tag{4.1.43}$$

Note however that $\nabla \times \mathbf{E}^{\perp} = \nabla \times \mathbf{E}$ since $\nabla \times \mathbf{E}^{||} = 0$ by definition. Therefore, we simply have

$$\nabla \times \mathbf{E} = -\partial_t \mathbf{B}. \tag{4.1.44}$$

This corresponds to Faraday's law in operator form.

We should mention that the electric and magnetic Gauss' laws follow as constraints in the quantization and were effectively imposed by setting the commutator of the displacement and magnetic field to be a transverse delta function. In the classical limit, we replace all operators with commuting numbers and we recover the classical Maxwell equations, suggesting the validity of our canonical quantization. We now move to study the eigenstates of the Hamiltonian, which are key to understanding quantum optics.

### 4.1.5 Photons in a medium

Let us consider what happens in one of the simplest possible cases: a linear, homogeneous, and isotropic medium. In this case, we can write the Maxwell equations for the electric fields as

$$\nabla^2 \mathbf{E} - \frac{n^2}{c^2} \partial_t^2 \mathbf{E} = 0. \tag{4.1.45}$$

It will be helpful to write the electric field as

$$\mathbf{E}(\mathbf{r}, t) = \int \frac{d^3k}{(2\pi)^3} \sum_{\lambda} E_{\lambda}(\mathbf{k}, t) e^{i\mathbf{k}\cdot\mathbf{r}} \hat{\epsilon}_{\lambda}(\mathbf{k}), \tag{4.1.46}$$

where $\hat{\epsilon}_{\lambda}(\mathbf{k})$ is a polarization vector satisfying $\mathbf{k} \cdot \hat{\epsilon}_{\lambda}(\mathbf{k}) = 0$. In other words, we are decomposing the electric field into a sum of transverse plane waves. We refer to this representation of the electric field as a "mode expansion". Now, recall that $\mathbf{E}$ is an operator, and it is an operator which represents a physically observable quantity and must therefore be Hermitian. That imposes a constraint:

$$E_{\lambda}(\mathbf{k}, t) \hat{\epsilon}_{\lambda}(\mathbf{k}) = E_{\lambda}(-\mathbf{k}, t)^{\dagger} \hat{\epsilon}^*_{\lambda}(-\mathbf{k}). \tag{4.1.47}$$

Note that the two-dimensional vector space spanned by the polarization vectors for $\mathbf{k}$ and $-\mathbf{k}$ is identical, and so we are free to impose for example that $\hat{\epsilon}_\lambda(\mathbf{k}) = \hat{\epsilon}^*_\lambda(-\mathbf{k})$. We can also impose orthonormality on the two polarization vectors for any given $\mathbf{k}$ such that $\hat{\epsilon}^*_\lambda(\mathbf{k}) \cdot \hat{\epsilon}_{\lambda'}(\mathbf{k}) = \delta_{\lambda\lambda'}$ (with $\lambda = 1, 2$). Therefore, we may simply say that Hermiticity requires

$$E_\lambda(\mathbf{k}, t) = E_\lambda(-\mathbf{k}, t)^\dagger. \tag{4.1.48}$$

Plugging our mode expansion into the operator Maxwell wave equation yields

$$\ddot{E}_\lambda(\mathbf{k}, t) + \omega_k^2 E_\lambda(\mathbf{k}, t) = 0, \tag{4.1.49}$$

with $\omega_k = ck/n$. The corresponding oscillator equation is solved by complex exponentials via

$$E_\lambda(\mathbf{k}, t) = a^E_\lambda(\mathbf{k}) e^{-i\omega_k t} + b^E_\lambda(\mathbf{k}) e^{i\omega_k t}. \tag{4.1.50}$$

The Hermiticity condition on $\mathbf{E}$ implies that

$$a^E_\lambda(\mathbf{k}) = b^E_\lambda(-\mathbf{k})^\dagger. \tag{4.1.51}$$

Therefore, we may write our mode expansion for the electric field operator as

$$\mathbf{E}(\mathbf{r}, t) = \int \frac{d^3k}{(2\pi)^3} \sum_\lambda \left( a^E_\lambda(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r} - i\omega_k t} \hat{\epsilon}_\lambda(\mathbf{k}) + a^E_\lambda(\mathbf{k})^\dagger e^{-i\mathbf{k}\cdot\mathbf{r} + i\omega_k t} \hat{\epsilon}^*_\lambda(\mathbf{k}) \right). \tag{4.1.52}$$

As of now, the $a_\lambda(\mathbf{k})$ operators are ostensibly arbitrary operator-valued constants. They however are not arbitrary as the corresponding $\mathbf{D}$ and $\mathbf{A}$ fields implied by this expression need to satisfy the canonical commutation relations. The $\mathbf{A}$ field is given by

$$\mathbf{A}(\mathbf{r}, t) = \int \frac{d^3k}{(2\pi)^3} \sum_\lambda \frac{1}{i\omega_k} \left( a^E_\lambda(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r} - i\omega_k t} \hat{\epsilon}_\lambda(\mathbf{k}) - a^E_\lambda(\mathbf{k})^\dagger e^{-i\mathbf{k}\cdot\mathbf{r} + i\omega_k t} \hat{\epsilon}^*_\lambda(\mathbf{k}) \right), \tag{4.1.53}$$

while the displacement field is given by

$$\mathbf{D}(\mathbf{r}, t) = \int \frac{d^3k}{(2\pi)^3} \sum_\lambda \epsilon_0 \epsilon \left( a^E_\lambda(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r} - i\omega_k t} \hat{\epsilon}_\lambda(\mathbf{k}) + a^E_\lambda(\mathbf{k})^\dagger e^{-i\mathbf{k}\cdot\mathbf{r} + i\omega_k t} \hat{\epsilon}^*_\lambda(\mathbf{k}) \right). \tag{4.1.54}$$

To evaluate the commutator $[D_i(\mathbf{r}), A_j(\mathbf{r}')]$, we will need to evaluate the commutator $[a^E_\lambda(\mathbf{k}), a^E_{\lambda'}(\mathbf{k}')^\dagger]$. To converge on final notation, let us define $a^E_\lambda(\mathbf{k}) = i\mathcal{E}_\lambda(\mathbf{k}) a_\lambda(\mathbf{k})$[7]. It is clear that from the standpoint of the electromagnetic field, different wavevectors are independent of each other and different polarizations are as well.

Therefore, we impose the commutation relation

$$[a^E_\lambda(\mathbf{k}), a^E_{\lambda'}(\mathbf{k}')^\dagger] \equiv |\mathcal{E}_\lambda(\mathbf{k})|^2 [a_\lambda(\mathbf{k}), a^\dagger_{\lambda'}(\mathbf{k}')] = |\mathcal{E}_\lambda(\mathbf{k})|^2 \times (2\pi)^3 \delta(\mathbf{k} - \mathbf{k}') \delta_{\lambda,\lambda'}. \tag{4.1.55}$$

[7]The factor of $i$ is arbitrary of course as we could have chosen any phase: we just choose the phase convention that aligns with standard texts on non-relativistic quantum electrodynamics.

Using these results, the equal-time[8] canonical commutator is found to be

$$[D_i(\mathbf{r},t),A_j(\mathbf{r},t)]=\int\frac{d^3k}{(2\pi)^3}\sum_\lambda|\mathcal{E}_\lambda(\mathbf{k})|^2\frac{i\epsilon_0\epsilon}{\omega_k}\left(e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')}\hat{\epsilon}_{\lambda,i}(\mathbf{k})\hat{\epsilon}^*_{\lambda,j}(\mathbf{k})+e^{-i\mathbf{k}(\mathbf{r}-\mathbf{r}')}\hat{\epsilon}^*_{\lambda,i}(\mathbf{k})\hat{\epsilon}_{\lambda,j}(\mathbf{k})\right). \tag{4.1.56}$$

If we take $|\mathcal{E}_\lambda(\mathbf{k})|^2=\frac{\hbar\omega_k}{2\epsilon_0\epsilon}$, we can perform the sum over polarizations and find[9]

$$[D_i(\mathbf{r},t),A_j(\mathbf{r},t)]=\frac{1}{2}i\hbar\int\frac{d^3k}{(2\pi)^3}(e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')}+e^{-i\mathbf{k}(\mathbf{r}-\mathbf{r}')})(\delta_{ij}-\hat{k}_i\hat{k}_j). \tag{4.1.57}$$

Taking $\mathbf{k}\to-\mathbf{k}$ in the second term immediately yields

$$[D_i(\mathbf{r},t),A_j(\mathbf{r},t)]=i\hbar\int\frac{d^3k}{(2\pi)^3}e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')}(\delta_{ij}-\hat{k}_i\hat{k}_j)=i\hbar\delta^\perp_{ij}(\mathbf{r}-\mathbf{r}'). \tag{4.1.58}$$

Summarizing, we may write our electric field operator as

$$\mathbf{E}(\mathbf{r},t)=i\int\frac{d^3k}{(2\pi)^3}\sum_\lambda\sqrt{\frac{\hbar\omega_k}{2\epsilon_0\epsilon}}\left(a_\lambda(\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{r}-i\omega_kt}\hat{\epsilon}_\lambda(\mathbf{k})-a_\lambda(\mathbf{k})^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}+i\omega_kt}\hat{\epsilon}^*_\lambda(\mathbf{k})\right). \tag{4.1.59}$$

The other operators follow immediately from the Maxwell equations as

$$\mathbf{D}(\mathbf{r},t)=i\int\frac{d^3k}{(2\pi)^3}\sum_\lambda\sqrt{\frac{\epsilon_0\epsilon\hbar\omega_k}{2}}\left(a_\lambda(\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{r}-i\omega_kt}\hat{\epsilon}_\lambda(\mathbf{k})-a_\lambda(\mathbf{k})^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}+i\omega_kt}\hat{\epsilon}^*_\lambda(\mathbf{k})\right) \tag{4.1.60}$$

and

$$\mathbf{A}(\mathbf{r},t)=\int\frac{d^3k}{(2\pi)^3}\sum_\lambda\sqrt{\frac{\hbar}{2\epsilon_0\epsilon\omega_k}}\left(a_\lambda(\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{r}-i\omega_kt}\hat{\epsilon}_\lambda(\mathbf{k})+a_\lambda(\mathbf{k})^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}+i\omega_kt}\hat{\epsilon}^*_\lambda(\mathbf{k})\right). \tag{4.1.61}$$

Let us use these results to express the Hamiltonian in terms of $a_\lambda(\mathbf{k})$ and $a_\lambda(\mathbf{k})^\dagger$. The result can be quickly found to be

$$H=\int\frac{d^3k}{(2\pi)^3}\sum_\lambda\frac{\hbar\omega_k}{2}\left(a_\lambda(\mathbf{k})a_\lambda(\mathbf{k})^\dagger+a_\lambda(\mathbf{k})^\dagger a_\lambda(\mathbf{k})\right) \tag{4.1.62}$$

Using the commutation relations for $a,a^\dagger$, we have

$$H=\int\frac{d^3k}{(2\pi)^3}\sum_\lambda\hbar\omega_ka(\mathbf{k})^\dagger a(\mathbf{k})+H_{\text{Z.P.}}, \tag{4.1.63}$$

[8] The commutators we have been writing thus far are Schrodinger picture operator commutators. In the Heisenberg picture, so long as the operators are evaluated at the same time, it is equal to the Schrodinger picture (t=0) commutator.

[9] I've used the fact that the two polarization vectors and $\hat{k}$ span three-dimensional space such that $\hat{k}_i\hat{k}_j+\sum_\lambda\hat{\epsilon}_{\lambda,i}(\mathbf{k})\hat{\epsilon}^*_{\lambda,j}(\mathbf{k})=\delta_{ij}$.

where $H_{\text{Z.P.}}$ is an infinite constant called the zero-point energy. It has no effect on physical predictions that we will make. Thus, in what follows, we will drop the constant and write

$$H = \int \frac{d^3k}{(2\pi)^3} \sum_\lambda \hbar\omega_k a(\mathbf{k})^\dagger a(\mathbf{k}). \tag{4.1.64}$$

Now, the commutation relations for $a, a^\dagger$ are clearly those of creation and annihilation operators of the harmonic oscillator. This should be no surprise as we essentially introduced these operators to diagonalize a quadratic Hamiltonian. The electromagnetic field Hamiltonian is essentially the field theory version of $p^2/2m + m\omega^2 x^2/2$.

### 4.1.6 Box quantization

While everything thus far has been rigorous, it is somewhat inconvenient to normalize $a, a^\dagger$ such that their commutation relations are delta functions. Instead, we consider our continuous space as the limit of a large box of volume $V$ with periodic boundaries as $V \to \infty$. The periodic boundary conditions impose that $\mathbf{k} = \frac{2\pi}{L}(m_x, m_y, m_z)$ where $m_{x,y,z}$ are integers. Then, our integrals over wavevectors are replaced by discrete sums and our delta-function commutators are replaced by unit commutators. Noting that $\sum_k = \frac{V}{(2\pi)^3}\int d^3k$, and that $\delta(\mathbf{k}=0) = \lim_{V\to 0} V/(2\pi)^3$, we may define a dimensionless $a_\mathbf{k} = a(\mathbf{k})/\sqrt{V}$ which has the property that $[a_{\mathbf{k},\lambda}, a^\dagger_{\mathbf{k}',\lambda'}] = \delta_{\mathbf{k},\mathbf{k}'}\delta_{\lambda,\lambda'}$. Then, we may write

$$H = \sum_{\mathbf{k},\lambda} \hbar\omega_k a^\dagger_{\mathbf{k},\lambda} a_{\mathbf{k},\lambda} + H_{\text{Z.P.}}, \tag{4.1.65}$$

with $[a_{\mathbf{k},\lambda}, a^\dagger_{\mathbf{k}',\lambda'}] = \delta_{\mathbf{k},\mathbf{k}'}\delta_{\lambda,\lambda'}$. Field operators may be written in the following way. The electric field operator can be written as

$$\mathbf{E}(\mathbf{r},t) = i\sum_{\mathbf{k},\lambda} \sqrt{\frac{\hbar\omega_k}{2\epsilon_0\epsilon V}} \left( a_{\mathbf{k},\lambda} e^{i\mathbf{k}\cdot\mathbf{r} - i\omega_k t}\hat{\epsilon}_{\mathbf{k},\lambda} - a^\dagger_{\mathbf{k},\lambda} e^{-i\mathbf{k}\cdot\mathbf{r} + i\omega_k t}\hat{\epsilon}^*_{\mathbf{k},\lambda} \right). \tag{4.1.66}$$

## 4.2 Quantum states of light

In the previous section, we showed that we could describe the electromagnetic field the way we would describe a collection of independent harmonic oscillators. There is one oscillator for each classical solution of Maxwell's equations, which in free space, is labeled by a wavevector and a polarization transverse to the wavevector. The electromagnetic field operators, specifically the vector potential and (minus) displacement field act as position and momentum-like variables and they are expressed in terms of linear combinations of creation and annihilation operators. The Hamiltonian, or energy, is also expressed in the way we expect for a collection of simple harmonic oscillators. Thus, we can now very easily talk about a basis of wavefunctions which span the Hilbert space describing electromagnetic field states. That Hilbert space, $\mathbb{H}_{\text{EM}}$ could be written as

$$\mathbb{H}_{\text{EM}} = \text{span}\{|n_{\mathbf{k}_1,\hat{\epsilon}_{\mathbf{k}_1}}, n_{\mathbf{k}_2,\hat{\epsilon}_{\mathbf{k}_2}}, \dots\rangle\} \equiv \text{span}\left\{ \bigotimes_{\mathbf{k},\hat{\epsilon}_\mathbf{k}} |n_{\mathbf{k},\hat{\epsilon}_\mathbf{k}}\rangle \right\}, \tag{4.2.1}$$

where $\otimes$ is the usual tensor product which allows us to adjoin multiple Hilbert spaces and the state $|n\rangle$ is a *number state*, *Fock state*, or simply, an *n-photon state*, defined via

$$|n\rangle = \frac{a^\dagger}{\sqrt{n}}|0\rangle, \tag{4.2.2}$$

with $|0\rangle$, the *zero-photon state* or *vacuum state* defined such that

$$a|0\rangle = 0. \tag{4.2.3}$$

The label $\mathbf{k}, \hat{\epsilon}_\mathbf{k}$ on $n_{\mathbf{k},\hat{\epsilon}_\mathbf{k}}$ denotes that we are taking about an $n$-photon state in a field mode with wavevector $\mathbf{k}$ and polarization $\hat{\epsilon}_\mathbf{k}$.

The notation is more complicated than the physics! The state $|n_{\mathbf{k}_1,\hat{\epsilon}_{\mathbf{k}_1}}, n_{\mathbf{k}_2,\hat{\epsilon}_{\mathbf{k}_2}}, \dots\rangle$ just represents a state in which there are $n_{\mathbf{k}_1,\hat{\epsilon}_{\mathbf{k}_1}}$ photons in $\mathbf{k}_1, \hat{\epsilon}_{\mathbf{k}_1}$, $n_{\mathbf{k}_2,\hat{\epsilon}_{\mathbf{k}_2}}$ photons in $\mathbf{k}_2, \hat{\epsilon}_{\mathbf{k}_2}$, $n_{\mathbf{k}_3,\hat{\epsilon}_{\mathbf{k}_3}}$ photons in $\mathbf{k}_3, \hat{\epsilon}_{\mathbf{k}_3}$ and so on. What the notation does however make abundantly clear is that the state space in quantum electrodynamics is vast in comparison with that of classical electrodynamics. In classical EM, the field is specified by one complex number for each wavevector-polarization pair. In quantum EM, each wavevector-polarization pair has an infinite-dimensional vector space attached to it! This much larger space of possibilities, directly leads to a number of behaviors of the EM field which are impossible to enable in classical EM. These novel behaviors typically concern the *statistical properties* of light. We will explore that in the following sections. To do this, let us take an *very* simple approach which is to consider an *single* mode of the electromagnetic field. Here, you can imagine a mode to be that which labels a classical solution of source-free Maxwell. In free-space, that is a wavevector-polarization pair. Other examples of single modes can be light confined in an optical cavity at a fixed resonance frequency. Although this may appear restrictive, many physically relevant situations—such as a Gaussian beam interacting with a nonlinear crystal—and sometimes even optical pulses, can be well understood at the qualitative level using the single-mode description.

### 4.2.1 Single-mode quantum states of light

A single mode of light is described by a quantum harmonic oscillator with Hamiltonian

$$H = \hbar\omega_0 a^\dagger a, \tag{4.2.4}$$

with $[a, a^\dagger] = 1$. The field operators in this case can be written for example as

$$\mathbf{E}(\mathbf{r}, t) = \mathcal{E}(\mathbf{r}) a e^{-i\omega t} + \mathcal{E}^*(\mathbf{r}) a^\dagger e^{i\omega t}. \tag{4.2.5}$$

For free space, $\mathcal{E}(\mathbf{r}) = \sqrt{\frac{\hbar\omega}{2\epsilon V}} e^{i\mathbf{k}\cdot\mathbf{r}} \hat{\epsilon}_\mathbf{k}$. For more complicated structured media, the $\mathcal{E}(\mathbf{r})$ satisfies the appropriate source-free Maxwell equation. We see that for a given position, the vector potential is proportional to $X \equiv (a + a^\dagger)/\sqrt{2}$ electric field is proportional to $P = i(a^\dagger - a)/\sqrt{2}$, which are position and momentum like operators of an oscillator. We refer to $X, P$ as *quadratures* of the electromagnetic field. They satisfy an uncertainty principle $\Delta X \Delta P = i$.

**Fock states**

We start our discussion of quantum states of light by analyzing simplest states from the Hamiltonian perspective, which are the eigenstates, called Fock states, or number states, or n-photon states. As you recall from your studies of the quantum harmonic oscillator[10] the states $|n\rangle = \frac{(a^\dagger)^n}{\sqrt{n!}}$ are eigenstates of the Hamiltonian with energy $E = n\hbar\omega$ (up to the overall constant). We can see why they're called n-photon states: we expect from our earliest interactions with wave-particle duality that $n$ photons will have energy $n\hbar\omega$ and we see that these n-photon states are labeled by non-negative integers. For this reason, we also call the operator $N = a^\dagger a$ the photon number operator, as its eigenstates are $n$-photon states $|n\rangle$ with eigenvalue $n$.

To get a better sense of these states, let us discuss expectation values of various operators:

1. The mean photon number $\langle N\rangle = \langle n|a^\dagger a|n\rangle = n\langle n|n\rangle = n$.

2. The variance in the photon number in the Fock state $|n\rangle$ is $(\Delta N)^2 = \langle N^2\rangle - \langle n\rangle^2 = 0$. This follows from the fact that for any operator function $f(N)$ we have $f(N)|n\rangle = f(n)$ and so $\langle N^2\rangle = n^2$.

3. The mean electric field at any point, $\langle E(\mathbf{r},t)\rangle$, is zero since $\langle n|a|n\rangle = \langle n|a^\dagger|n\rangle^* = 0$. Similarly, since the magnetic field is also a linear combination of $a, a^\dagger$ its mean is also zero.

4. The variance in any component of the electric field is non-zero. Since the mean vanishes, we have $(\Delta E_i(\mathbf{r},t))^2 = \langle E_i^2(\mathbf{r},t)\rangle = \langle n|\left(\mathcal{E}_i^2(\mathbf{r})a^2e^{-2i\omega t} + |\mathcal{E}_i(\mathbf{r})|^2(aa^\dagger + a^\dagger a) + \mathcal{E}_i^{*2}(\mathbf{r})a^{\dagger 2}e^{2i\omega t}\right)|n\rangle$.

[10] This footnote provides a short refresher on oscillator eigenstates. The quantum harmonic oscillator admits a basis of states with well-defined photon number, known as Fock states, denoted $|n\rangle$. These states have an exactly defined number of photons in the mode. For example, $|0\rangle$ contains zero photons and is referred to as the vacuum state, while $|1\rangle$ is a state containing a single photon. The creation and annihilation operators act on these Fock states by creating and destroying photons, with the algebraic rules:

$$a\,|n\rangle = \sqrt{n}\,|n-1\rangle \tag{4.2.6}$$

$$a^\dagger\,|n\rangle = \sqrt{n+1}\,|n+1\rangle\,. \tag{4.2.7}$$

For example, the single photon Fock state is $|1\rangle = a^\dagger\,|0\rangle$. Higher order Fock states can be created by applying the creation operator multiple times. As a result, a Fock state with $n$ photons is generally given by

$$|n\rangle = \frac{(a^\dagger)^n}{\sqrt{n!}}\,|0\rangle\,. \tag{4.2.8}$$

Fock states are often discussed in the context of the number operator:

$$N \equiv a^\dagger a. \tag{4.2.9}$$

Specifically, Fock states $|n\rangle$ are eigenstates of $n$

$$N\,|n\rangle = n\,|n\rangle\,, \quad n = 0, 1, 2, \ldots \tag{4.2.10}$$

They also form an orthonormal basis for the so-called "Hilbert space" of a quantum oscillator mode:

$$\langle m|n\rangle = \delta_{mn}, \qquad \sum_{n=0}^{\infty} |n\rangle\langle n| = I. \tag{4.2.11}$$

The terms with $a^2, a^{\dagger 2}$ give no contribution. The remain terms can be evaluated using $aa^\dagger + a^\dagger a = 2N + 1$, leaving us with $(\Delta E_i(\mathbf{r}, t))^2 = (2n + 1)|\mathcal{E}_i(\mathbf{r})|^2$.

5. It is useful to also talk about the quadrature variances. It is easy to see that $(\Delta X)^2 = (\Delta P)^2 = n + \frac{1}{2}$.

The first two properties are expected from the name: an $n$-photon state should have a well-defined number of photons $n$ and so there should be zero variance around the mean $n$. The electric field is less intuitive, and tells you already that the Fock state is not like the lasers we work with in optics typically, that have a well-defined average, sinusoidally oscillating field. The average field is zero, but its square is nonzero on average - representing a type of light with a random electric field [11].

Importantly, the field variance is nonzero even for the ground state of the harmonic oscillator, which has no photons in it! This nonzero variance in the absence of photons is known as vacuum fluctuations (or zero-point fluctuations). It reflects the fact that the electric field, like position in a harmonic oscillator, cannot be sharply defined in the ground state due to the uncertainty principle. We highlight that this is a purely quantum phenomenon with no classical analog: in classical electromagnetism, a field with zero amplitude has no fluctuations and carries no energy. In contrast, the quantum vacuum is an active state, with inevitable fluctuations which arise from quantum mechanical uncertainty. These vacuum fluctuations play a central role in many phenomena in quantum and nonlinear optics. For example, they act as the seed for spontaneous processes such as spontaneous emission and parametric down-conversion, and set fundamental noise limits in optical measurements. More broadly, they provide the background upon which nonlinear interactions generate nonclassical states of light, such as squeezed and entangled states, which we will study in later sections.

### 4.2.2 Coherent states

Let us now discuss a class of states which are in some sense, the "most classical states" of the electromagnetic field. These states have a well-defined expectation value of the electromagnetic field which satisfies the classical Maxwell equations, and in the limit of a large enough number of photons, has minimal relative fluctuations of the field relative to the average. In other words, they represent a classical electromagnetic field with a small amount of quantum noise on top, which can be thought of as vacuum fluctuations. These states are called *coherent states*, are labeled $|\alpha\rangle$ and can be defined in the following way:

$$|\alpha\rangle = D(\alpha)|0\rangle \equiv e^{\alpha a^\dagger - \alpha^* a}|0\rangle, \tag{4.2.12}$$

where $D(\alpha)$ is a unitary operator called the *displacement operator*. A useful way to calculate expectation values is in terms of a Heisenberg picture. Although we are not explicitly referring

[11] For the $n$ photon state, these properties can be understood by analogy to the harmonic oscillator in the coordinate representation. We know that the electric field operator is like the $p$ operator, and so we can talk about the momentum wavefunctions of the oscillator eigenstates. You may be less familiar with the momentum wavefunctions of the eigenstates, but they are the same as the position wavefunctions since the Hermite-Gaussian polynomials are their own Fourier transform. Therefore, we can say there is a wavefunction whose square gives the probability of measuring the electric field at each point and it is given by a Hermite-Gaussian.

to time, since $D(\alpha)$ is unitary, we can talk about unitary as affecting a corresponding operator transformation

$$a \to D^\dagger(\alpha) a D(\alpha). \tag{4.2.13}$$

The most standard way to evaluate this is using the so-called Campbell identity[12]. The Campbell identity applied in this context,with $B = a, A = \alpha^* a - \alpha a^\dagger$, states that

$$D^\dagger(\alpha) a D(\alpha) = a + \alpha. \tag{4.2.15}$$

All higher-order terms in the series vanish. We see that the displacement operator indeed displaces the annihilation operator. Let us now ask about some of the expectation values we discussed before, but in reverse order. Let's talk about fields, and then photon numbers. Let us also ask about what happens when the coherent state freely evolves in time. If we have a coherent state at time $t = 0$, then at time $t$, the resulting state is

$$|\alpha(t)\rangle = e^{-iHt/\hbar}|\alpha\rangle = e^{-i\omega t N}|\alpha\rangle = e^{-i\omega t N} D(\alpha)|0\rangle. \tag{4.2.16}$$

To evaluate this, we multiply by one:

$$|\alpha(t)\rangle = e^{-iHt/\hbar}|\alpha\rangle = e^{-i\omega t N} D(\alpha) e^{i\omega t N} e^{-i\omega t N}|0\rangle. \tag{4.2.17}$$

Using $e^{-i\omega t N} a e^{i\omega t N} = a e^{-i\omega t}$ and $N|0\rangle = 0$, we may write[13]

$$|\alpha(t)\rangle = D(\alpha e^{-i\omega t})|0\rangle. \tag{4.2.18}$$

Therefore the operator at time $t$ may be written as

$$a(t) = a + \alpha e^{-i\omega t}. \tag{4.2.19}$$

With this we may now straightforwardly compute various expectation values. Let's start with the $i$th component of the field. In this Heisenberg picture, this is computed as

$$\langle E_i(r,t)\rangle = \langle \mathcal{E}(\mathbf{r}) a(t) + \mathcal{E}^*(\mathbf{r}) a^\dagger(t)\rangle = \langle 0|\mathcal{E}(\mathbf{r}) a(t) + \mathcal{E}^*(\mathbf{r}) a^\dagger(t)|0\rangle. \tag{4.2.20}$$

Importantly, the expectation value is taken with respect to the initial state, which can be taken as zero if we treat $a(t)$ as resulting from the combination of displacement and unitary time evolution. Therefore the answer is immediately found to be

$$\mathcal{E}(\mathbf{r})\alpha(t) e^{-i\omega t} + \mathcal{E}^*(\mathbf{r}) a^* e^{i\omega t}. \tag{4.2.21}$$

[12]The Campbell identity is a useful formula often used to evaluate unitary transformations in quantum mechanics. Its statement is: consider two operators $A$ and $B$ and consider the quantity $e^A B e^{-A}$. Then it is given by

$$e^A B e^{-A} = B + [A,B] + \frac{1}{2!}[A,[A,B]] + \frac{1}{3!}[A,[A,[A,B]]] + \dots. \tag{4.2.14}$$

A corollary of this identity is that if $[A,B] = cI$ with $I$ the identity, then $e^A B e^{-A} = B + c$.

[13]As a reminder, in the Heisenberg picture, the free evolution of $a$, given by $a(t) = U^\dagger a U$ with $U = e^{-iHt/\hbar}$ is given by $\dot{a} = \frac{i}{\hbar}[\hbar\omega a^\dagger a, a] = -i\omega a \implies a(t) = e^{-i\omega t} a(0)$. Further, for any operator like $D(\alpha)$, we can see that by expanding it in a Taylor series: $D(\alpha) = \sum_n \frac{1}{n!}(\alpha a^\dagger - \alpha^* a)^n$, the Heisenberg evolution of such an operator just amounts to replacing $a \to a(t)$ and $a^\dagger \to a^\dagger(t)$.

This is finite and corresponds to a sinusoidal oscillation of the field, like you might imagine classically. The variance is nonzero, and is given by

$$(\Delta E_i(r,t))^2 = |\mathcal{E}(\mathbf{r})|^2, \tag{4.2.22}$$

which is is time-independent, and exactly the same value as in the vacuum state. Similarly to the Fock states, it is helpful to talk about the quadrature variances, which are easily found as $(\Delta X)^2 = (\Delta P)^2 = 1/2$.

The interpretation here is that the coherent state is a sinusoidal field with small vacuum or zero-point fluctuations. It may be intuitively thought of as having the least amount of noise possible (but this is not exactly right, as it is possible to find states with field variances below the value mentioned here).

**Schrodinger picture description of coherent states**

Let us also discuss the coherent state in its Fock representation (or equivalently, its energy eigenbasis representation).

We saw previously that Fock states have peculiar statistical properties (such as zero average electric field) which do not connect well to our classical conception of electromagnetism. It is thus natural to wonder about what quantum state of light might behave like a classical optical field. Such states are known as "coherent states," which are the topic of this section.

The coherent state can be seen as resulting from a unitary transformation of the vacuum state. We are then tasked with evaluating

$$|\alpha\rangle = D(\alpha)|0\rangle \equiv e^{\alpha a^\dagger - \alpha^* a}|0\rangle. \tag{4.2.23}$$

The simplest way to do this is to note that the transformation $D^\dagger(\alpha) a D(\alpha) = a + \alpha$ implies that the coherent state is an eigenstate of the annihilation operator. To see this, consider

$$a|\alpha\rangle = DD^\dagger a D|0\rangle = D(a+\alpha)|0\rangle = \alpha|\alpha\rangle. \tag{4.2.24}$$

If we express an eigenstate of the annihilation operator in the energy / photon number eigenbasis, as

$$|\alpha\rangle = \sum_{n=0}^{\infty} c_n |n\rangle, \tag{4.2.25}$$

then the eigenstate condition implies

$$a|\alpha\rangle = \sum_{n=0}^{\infty} c_n \sqrt{n}|n-1\rangle = \sum_{n=0}^{\infty} c_{n+1}\sqrt{n+1}|n\rangle = \alpha \sum_{n=0}^{\infty} c_n |n\rangle, \tag{4.2.26}$$

implying the recurrence relation $c_{n+1} = \frac{\alpha}{\sqrt{n+1}} c_n$.Taking $c_0 = \mathcal{N}$ with $\mathcal{N}$ a normalization constant implies

$$|\alpha\rangle = \mathcal{N} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}}|n\rangle. \tag{4.2.27}$$

The normalization condition is $|\mathcal{N}|^2 \sum_{n=0}^{\infty} \frac{|\alpha|^{2n}}{n!} = 1$ which implies $|\mathcal{N}| = e^{-\frac{1}{2}|\alpha|^2}$. This allows us to write the coherent state in the Fock basis as

$$|\alpha\rangle = e^{-\frac{1}{2}|\alpha|^2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle. \tag{4.2.28}$$

Importantly, the coherent state is not an eigenstate of the Hamiltonian and is a superposition of photon number states. The probability of finding the system to have $n$ photons is

$$P(n) = e^{-|\alpha|^2} \frac{|\alpha|^{2n}}{n!}, \tag{4.2.29}$$

which is a Poisson distribution of mean $\langle n \rangle = |\alpha|^2$. The variance of the photon number is equal to the mean (and all cumulants of the photon number distribution generally are equal).

### 4.2.3 Squeezed states

We saw in the previous section that coherent states exhibit vacuum fluctuations: while they have a nonzero mean electric field, their fluctuations are the same as those of the vacuum. In particular, the uncertainty principle constrains the product of quadrature variances, fixing a lower bound on how small the fluctuations can be. This naturally raises the question of whether it is possible to reduce the fluctuations in one quadrature below the vacuum level, at the expense of increased fluctuations in the conjugate quadrature. The class of states known as squeezed states exhibits precisely this behavior.

To introduce squeezed states, we define the single-mode squeezing operator:

$$S(\zeta) = \exp\left(\frac{1}{2}\zeta^* a^2 - \frac{1}{2}\zeta (a^\dagger)^2\right), \tag{4.2.30}$$

where

$$\zeta = r e^{i\phi} \tag{4.2.31}$$

is the complex squeezing parameter. While this may seem unmotivated at the moment, we note that this form of the squeezing operator arises naturally from common interactions in nonlinear optics, such as parametric amplification in $\chi^{(2)}$ media or four-wave mixing in $\chi^{(3)}$ systems, where pairs of photons are created or annihilated. We will show this in greater detail later.

A state known as the "squeezed vacuum" state is obtained by applying $S(\zeta)$ to the vacuum:

$$|\zeta\rangle = S(\zeta)\,|0\rangle\,. \tag{4.2.32}$$

A key property is that $S(\zeta)$ performs a Bogoliubov transformation on the operators:

$$S^\dagger(\zeta)\, a\, S(\zeta) = a \cosh r - e^{i\phi} a^\dagger \sinh r, \tag{4.2.33}$$

$$S^\dagger(\zeta)\, a^\dagger\, S(\zeta) = a^\dagger \cosh r - e^{-i\phi} a \sinh r. \tag{4.2.34}$$

In what follows, we will write the transformation as

$$a \to \mu a + \nu a^\dagger, \tag{4.2.35}$$

where $\mu = \cosh r, \nu = -e^{i\phi}$.

We now compute basic expectation values in the squeezed vacuum state. First, the mean field vanishes:

$$\langle a \rangle = \langle \zeta | \, a \, | \zeta \rangle \tag{4.2.36}$$

$$= \langle 0 | \, S^\dagger(\zeta) \, a \, S(\zeta) \, | 0 \rangle \tag{4.2.37}$$

$$= \langle 0 | \, \mu a + \nu a^\dagger \, | 0 \rangle = 0, \tag{4.2.38}$$

and similarly

$$\langle a^\dagger \rangle = 0. \tag{4.2.39}$$

This implies that any quadrature has mean value zero and therefore the electric and magnetic fields are also on average zero.

The mean photon number can be computed by explicitly using $|\zeta\rangle = S(\zeta)\,|0\rangle$:

$$\langle n \rangle = \langle \zeta | \, a^\dagger a \, | \zeta \rangle \tag{4.2.40}$$

$$= \langle 0 | \, S^\dagger(\zeta) \, a^\dagger a \, S(\zeta) \, | 0 \rangle \tag{4.2.41}$$

$$= \langle 0 | \, (\mu^* a^\dagger + \nu^* a)(\mu a + \nu a^\dagger) \, | 0 \rangle \tag{4.2.42}$$

$$= |\nu|^2 = \sinh^2 r. \tag{4.2.43}$$

This is not zero, which is to be expected since the squeezed state is not a Hamiltonian eigenstate and thus must be a superposition of different photon number states. When $r \gg 1$, we see that the number of photons goes like $e^{2r}/4$ indicating a type of exponential gain[14].

Let us now look at the quadrature variances associated with the squeezed state. The variance

$$(\Delta X_\theta)^2 = \frac{1}{2} \langle 0 | \left( (\mu a + \nu a^\dagger) e^{-i\theta} + (\mu^* a^\dagger + \nu^* a) e^{i\theta} \right)^2 | 0 \rangle \tag{4.2.44}$$

$$= \frac{1}{2} \langle 0 | \left( (\mu e^{-i\theta} + \nu^* e^{i\theta}) a + (\mu^* e^{i\theta} + \nu e^{-i\theta}) a^\dagger \right)^2 | 0 \rangle \tag{4.2.45}$$

$$= \frac{1}{2} \left| \mu e^{-i\theta} + \nu^* e^{i\theta} \right|^2 \tag{4.2.46}$$

$$= \frac{1}{2} \left| \cosh r e^{-i\theta} - \sinh r e^{i\theta - i\phi} \right|^2 \tag{4.2.47}$$

$$= \frac{1}{2} \left| \cosh r e^{-i\psi} - \sinh r e^{i\psi} \right|^2, \tag{4.2.48}$$

where I have defined $\psi = \theta - \phi/2$.

Let us consider the quadrature angle $\theta$ that maximizes and minimizes the variance. Since the complex exponentials have magnitude one, the maximum and minimum values correspond to

$$(\Delta X_\theta)^2_{\max} = \frac{1}{2} e^{2r}, (\Delta X_\theta)^2_{\min} = \frac{1}{2} e^{-2r}. \tag{4.2.49}$$

[14] It is no accident that this looks very similar to the intensity of what is generated by a parametric amplifier. In fact, you may remember when we studied the degenerate parametric amplifier, we found a classical field transformation of the form $A(0) \to A(0)\cosh r + A^*(0) e^{i\phi} \sinh r$. We also found that the intensity for large $r$ goes like $e^{2r}/4$. The Bogoliubov transformation is exactly the linear operator transformation corresponding to this classical transformation and it would therefore should not surprise you that the way to produce a squeezed state is by sending vacuum into a degenerate parametric amplifier!

The angles that lead to minimum and maximum variance are separated by $\pi/2$ and thus have the property that

$$(\Delta X_\theta)^2_{\max}(\Delta X_\theta)^2_{\min} = \frac{1}{4}. \tag{4.2.50}$$

Compared to the vacuum state and the coherent state, the variance along some quadratures are reduced relative to the vacuum level at the expense of enhanced fluctuations in other quadratures. This is the defining property of squeezing. The reduced variance in a particular quadrature enables stronger sensitivity in metrology when we are measuring a weak signal which is encoded in that particular quadrature. The property of having a variance which is reduced compared to the vacuum level is a quantum mechanical effect and cannot be reproduced by any classical random electromagnetic field.

So far, we have considered squeezed states generated from acting the squeezing operator $S(\zeta)$ on the vacuum state. These states have zero mean fields, but nonzero variances as we have just described. More generally, one can consider a larger family of squeezed states which are obtained by displacing squeezed vacuum states.

Thie can be done by defining a displaced squeezed state:

$$|\alpha, \zeta\rangle = D(\alpha)S(\zeta)\,|0\rangle\,. \tag{4.2.51}$$

Such a state has average properties given by the coherent displacement:

$$\langle\alpha, \zeta|a|\alpha, \zeta\rangle = \alpha, \qquad \langle\alpha, \zeta|a^\dagger|\alpha, \zeta\rangle = \alpha^*, \tag{4.2.52}$$

but retains the quadrature variance properties associated with the squeezed state. Such states are sometimes referred to as "bright squeezed states" since they have a nonzero average field. This is in contrast to squeezed vacuum states, which contain few photons on average.

### 4.2.4 (Optional) Derivation of Bogoliubov transformation on oscillator operators

In this optional section, we derive the Bogoliubov transformation generated by the single-mode squeezing operator. We begin with the definition:

$$S(\zeta) = \exp\left(\frac{1}{2}\zeta^* a^2 - \frac{1}{2}\zeta(a^\dagger)^2\right), \qquad \zeta = re^{i\phi}. \tag{4.2.53}$$

We define:

$$G \equiv \frac{1}{2}\zeta^* a^2 - \frac{1}{2}\zeta(a^\dagger)^2, \tag{4.2.54}$$

so that

$$S(\zeta) = e^G. \tag{4.2.55}$$

Our objective is to compute a transformation of the annihilation operator:

$$S^\dagger(\zeta)\, a\, S(\zeta) = e^{-G} a e^G. \tag{4.2.56}$$

We will use the BCH expansion,

$$e^{-G} a e^G = a + [a, G] + \frac{1}{2!}[[a, G], G] + \frac{1}{3!}[[[a, G], G], G] + \cdots \tag{4.2.57}$$

We first compute the needed commutators. Using $[a, a^\dagger] = 1$,

$$[a, a^2] = 0, \tag{4.2.58}$$

and

$$\begin{aligned}
[a, (a^\dagger)^2] &= [a, a^\dagger]a^\dagger + a^\dagger[a, a^\dagger] && (4.2.59)\\
&= a^\dagger + a^\dagger && (4.2.60)\\
&= 2a^\dagger. && (4.2.61)
\end{aligned}$$

Therefore,

$$\begin{aligned}
[a, G] &= \left[a, \frac{1}{2}\zeta^* a^2 - \frac{1}{2}\zeta(a^\dagger)^2\right] && (4.2.62)\\
&= \frac{1}{2}\zeta^*[a, a^2] - \frac{1}{2}\zeta[a, (a^\dagger)^2] && (4.2.63)\\
&= -\zeta a^\dagger. && (4.2.64)
\end{aligned}$$

Next,

$$\begin{aligned}
[[a, G], G] &= [-\zeta a^\dagger, G] && (4.2.65)\\
&= -\zeta[a^\dagger, G]. && (4.2.66)
\end{aligned}$$

Now

$$[a^\dagger, (a^\dagger)^2] = 0, \tag{4.2.67}$$

and

$$\begin{aligned}
[a^\dagger, a^2] &= [a^\dagger, a]a + a[a^\dagger, a] && (4.2.68)\\
&= -a - a && (4.2.69)\\
&= -2a. && (4.2.70)
\end{aligned}$$

So

$$\begin{aligned}
[a^\dagger, G] &= \left[a^\dagger, \frac{1}{2}\zeta^* a^2 - \frac{1}{2}\zeta(a^\dagger)^2\right] && (4.2.71)\\
&= \frac{1}{2}\zeta^*[a^\dagger, a^2] - \frac{1}{2}\zeta[a^\dagger, (a^\dagger)^2] && (4.2.72)\\
&= -\zeta^* a. && (4.2.73)
\end{aligned}$$

Hence

$$[[a, G], G] = |\zeta|^2 a. \tag{4.2.74}$$

Continuing once more,

$$\begin{aligned}
[[[a, G], G], G] &= [|\zeta|^2 a, G] && (4.2.75)\\
&= |\zeta|^2[a, G] && (4.2.76)\\
&= -|\zeta|^2\zeta a^\dagger. && (4.2.77)
\end{aligned}$$

The pattern is now clear:

$$\text{odd nested commutators} \propto a^\dagger, \tag{4.2.78}$$

$$\text{even nested commutators} \propto a. \tag{4.2.79}$$

Substituting into the BCH series gives

$$e^{-G}ae^{G} = a - \zeta a^\dagger + \frac{|\zeta|^2}{2!}a - \frac{|\zeta|^2\zeta}{3!}a^\dagger + \frac{|\zeta|^4}{4!}a - \cdots \tag{4.2.80}$$

$$= \left(1 + \frac{|\zeta|^2}{2!} + \frac{|\zeta|^4}{4!} + \cdots\right)a - \left(\zeta + \frac{|\zeta|^2\zeta}{3!} + \frac{|\zeta|^4\zeta}{5!} + \cdots\right)a^\dagger. \tag{4.2.81}$$

Writing $\zeta = re^{i\phi}$ with $|\zeta| = r$, this becomes

$$e^{-G}ae^{G} = \left(1 + \frac{r^2}{2!} + \frac{r^4}{4!} + \cdots\right)a - e^{i\phi}\left(r + \frac{r^3}{3!} + \frac{r^5}{5!} + \cdots\right)a^\dagger \tag{4.2.82}$$

$$= a\cosh r - e^{i\phi}a^\dagger \sinh r. \tag{4.2.83}$$

Thus,

$$S^\dagger(\zeta)\, a\, S(\zeta) = a\cosh r - e^{i\phi}a^\dagger \sinh r. \tag{4.2.84}$$

Taking the Hermitian conjugate gives the companion relation

$$S^\dagger(\zeta)\, a^\dagger\, S(\zeta) = a^\dagger \cosh r - e^{-i\phi}a \sinh r. \tag{4.2.85}$$

This is the Bogoliubov transformation associated with single-mode squeezing. This mixing of creation and annihilation operators is typical of nonlinear optical interactions, as we will continue to see in later sections.

## 4.3 Scattering and attenuation of light

Thus far, we have studied quantum states in isolation of all other influences. In other words, we constructed different quantum states of light (Fock states, coherent states, squeezed states) and analyzed their properties and the only dynamics we considered was their evolution under the Hamiltonian. In this section, we consider what happens when quantum light undergoes a beamsplitting operation. Although this sounds very specific, it turns out an analysis of a beamsplitter provides a general quantum-mechanically consistent framework for describing the attenuation of light, which, if done naively, violates the laws of quantum mechanics! The beamsplitter has some other beautiful and important properties. For example, it implements the displacement operation, and it also tells us that certain types of intricate quantum correlations such as squeezing are degraded by attenuation, which illustrates one of the central problems in the productive application of quantum optics. The beamsplitter analysis also immediately furnishes a theory of reflection and transmission.

In classical optics, it is common to model the behavior of a beamsplitter as a linear transformation between input and output fields. In particular, if $\alpha_{1,2}$ are the complex electric

fields at the inputs, and $\beta_{1,2}$ are the complex electric fields at the outputs, then we can write[15]:

$$\begin{pmatrix}\beta_1\\ \beta_2\end{pmatrix} = \begin{pmatrix}-r & it\\ it & -r\end{pmatrix}\begin{pmatrix}\alpha_1\\ \alpha_2\end{pmatrix}. \tag{4.3.1}$$

Here, $r$ and $t$ are complex reflection and transmission coefficients. The classical power between the two ports must be conserved. This puts a constraint on the values of the reflection and transmission coefficients. In particular, the matrix which transforms the fields must be unitary. This means

$$\begin{pmatrix}-r^* & -it^*\\ -it^* & -r^*\end{pmatrix}\begin{pmatrix}-r & it\\ it & -r\end{pmatrix} = \begin{pmatrix}|t|^2+|r|^2 & -itr^*+it^*r\\ it^*r-itr^* & |t|^2+|r|^2\end{pmatrix} = \begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix}. \tag{4.3.2}$$

The two independent equations that $r$ and $t$ must satisfy are thus:

$$|r|^2+|t|^2 = 1 \tag{4.3.3}$$

$$rt^* - r^*t = 0. \tag{4.3.4}$$

The first constraint comes directly from power conservation, while the second can be seen a reciprocity condition. The exact values of $r$ and $t$ depend on the details of the physical implementation. If the beamsplitter consists of interfaces between different dielectric layers, then they can be found by solving a boundary condition problem for the fields in various layers.

Now we see that the beamsplitter induces a linear transformation on the classical fields. We may thus expect that this is also a valid *operator transformation* in the Heisenberg sense. In other words, we may think of $a_1, a_2$ as input field operators which get *transformed* by the beamsplitter to output field operators $b_1$ and $b_2$. Importantly, if the transformation is to be a valid Heisenberg transformation, then it must conserve all the relevant commutators. In other words, if $[a_i, a_j^\dagger] = \delta_{ij}$ then $[b_i, b_j^\dagger] = \delta_{ij}$. Let us denote the matrix relating $(a_1, a_2)$ to $(b_1, b_2)$ as $S$ such that

$$\begin{pmatrix}b_1\\ b_2\end{pmatrix} = S\begin{pmatrix}a_1\\ a_2\end{pmatrix}. \tag{4.3.5}$$

Then we may evaluate

$$[b_i, b_j^\dagger] = [S_{i1}a_1 + S_{i2}a_2, S_{j1}^*a_1^\dagger + S_{j2}^*a_2^\dagger] = S_{i1}S_{j1}^* + S_{i2}S_{j2}^* = \delta_{ij}. \tag{4.3.6}$$

In other words, the $i$ and $j$ rows must be orthonormal vectors, implying that $S$ is unitary. Therefore, we may say that our beamsplitter, in the Heisenberg picture, implements a unitary matrix on the input operators and maps them to output operators. Let us now explore some of the consequences of this simple relationship.

### 4.3.1 Displacement operator

Let us consider a situation in which there is a quantum state incident on a first port described by annihilation operator $a_1$ while in the second port we have a coherent state $|\alpha\rangle$ incident on

[15]There are multiple valid conventions for the S-matrix. I am using the one in Waves and Fields in Optoelectronics by Haus.

the second port described by operator $a_2$. For a strong coherent state in the second mode, we know that

$$a_2 = \alpha + \delta a_2, \tag{4.3.7}$$

where $\delta a_2$ represents the Heisenberg operator prior to displacement [16]. When taking expectation values involving $a_2$ they are now to be taken against the vacuum state (while they would have been taken against the coherent state had we simply worked with $a_2$). Now, let us consider the case when $|t|^2 \ll 1$ and $|t\alpha| \gg 1$ (meaning that the number of photons transmitted from port 2 is much larger than one). Then we may write

$$b_1 = -ra_1 + it(\alpha + \delta a_2). \tag{4.3.8}$$

Since $|r| \approx 1$ and $|t| \ll 1$, we see that in general $t\delta a_2$ will have negligible contribution to expectation values when $t \to 0$. Therefore, we may approximate

$$b_1 \approx \beta_1 - ra_1, \tag{4.3.9}$$

where $\beta = it\alpha_1$. Thus, we see that $a_1$ is displaced, up to an overall phase. In particular, taking $r = |r|e^{i\phi}$ and approximating $|r| \approx 1$, we have

$$b_1 \approx \beta_1 - e^{i\phi}a_1. \tag{4.3.10}$$

We thus see that the beamsplitter acts as the displacement operator!

### 4.3.2 Beamsplitter as attenuation

Consider the case in which the first port has a state $|\psi\rangle$ incident on it and the second port has nothing incident on it. Quantum mechanics tells us that nothing does not really exist, and that we should think of the second port as having $|0\rangle$ incident on it. Let us compute how expectation values of various operators change as a result of this. Consider first the photon number operator

$$\langle b_1^\dagger b_1\rangle = \langle(-r^*a_1^\dagger - it^*a_2^\dagger)(-ra_1 + ita_2)\rangle = |r|^2\langle a_1^\dagger a_1\rangle. \tag{4.3.11}$$

This is exactly what we intuitively expect. The energy incident in the first port is attenuated to a factor $|r|^2$. Quadrature operators in their expectation values also transform roughly in the way you expect

$$\langle X_{\theta,\text{out}}\rangle = \left\langle \frac{b_1 e^{-i\theta} + b_1^\dagger e^{i\theta}}{\sqrt{2}} \right\rangle = \frac{1}{\sqrt{2}}\left(-ra_1 e^{-i\theta} - r^* a_1^\dagger e^{i\theta}\right) = -|r|\langle X_{\theta-\phi,,\text{in}}\rangle, \tag{4.3.12}$$

where we have written $r = |r|e^{i\phi}$ and used the notations "in" and "out" to denote which operators are computed before and after the beamsplitter, respectively. There is a rotation of the quadrature which depends on the complex phase of the reflection coefficient.

[16] The notation $\delta$ seems a bit odd at this point. We will make further use of it. But you should think of it as saying: the operator can be seen as mostly a c-number plus a quantum noise associated with the vacuum state.

Thus far, the fact that vacuum is incident on the second port of the beamsplitter has not worked its way in at all, and that is rather intuitive. Perhaps less intuitive is what happens to quadrature variances. Let us consider the expectation value of

$$\langle X^2_{\theta,\text{out}}\rangle = \frac{1}{2}\Big\langle \left(b_1^2 e^{-2i\theta} + b_1^{\dagger 2} e^{2i\theta} + b_1 b_1^\dagger + b_1^\dagger b_1\right)\Big\rangle. \tag{4.3.13}$$

The first term is $-\frac{1}{2}r^2\langle a_1^2\rangle e^{-2i\theta}$, while the second term is $-\frac{1}{2}r^{*2}\langle a_1^{\dagger 2}\rangle e^{2i\theta}$. The third and fourth terms are most quickly evaluated by expressing their sum as $2\langle b_1^\dagger b_1\rangle + 1 = 2|r|^2\langle a_1^\dagger a_1\rangle + 1$.

We may now write the variance as

$$(\Delta X_{\theta,\text{out}})^2 = \frac{|r|^2}{2}\left(\langle a_1^2\rangle e^{-2i(\theta-\phi)} + \langle a_1^{\dagger 2}\rangle e^{2i(\theta-\phi)} + 2\langle a_1^\dagger a_1\rangle\right) + \frac{1}{2} - |r|^2\langle X_{\theta-\phi,\text{in}}\rangle^2. \tag{4.3.14}$$

To get this into a cleaner form, we note that

$$X^2_{\theta-\phi,\text{in}} = \frac{a_1^2 e^{-2i(\theta-\phi)} + a_1^{\dagger 2} e^{+2i(\theta-\phi)} + 2a_1^\dagger a_1 + 1}{2}, \tag{4.3.15}$$

which allows us to write

$$(\Delta X_{\theta,\text{out}})^2 = |r|^2(\Delta X_{\theta-\phi,\text{in}})^2 + \frac{1}{2}|t|^2, \tag{4.3.16}$$

where I have used $|r|^2 + |t|^2 = 1$. Naively, one might have expected that the attenuation by $r$ would merely scale the quadrature variance by $|r|^2$ but we see that there is an other term which appears. This term makes the limit of $|r| \to 0$ physically sensible. For example, in the limit of complete attenuation, we expect to get vacuum which has variance of $1/2$ in any quadrature. This is precisely what happens! If we did not include this term, we would have violated the uncertainty principle because we would have $\Delta X_\theta \Delta X_{\theta+\pi/2} < 1/2$.

An interesting consequence of this expression is the degradation of squeezing. Suppose without loss of generality that the reflection phase is $\phi = 0$ and we have light whose $\theta$ quadrature is squeezed such that $(\Delta X_{\theta,\text{in}})^2 = \frac{1}{2}\epsilon$ with $\epsilon < 1$. Then

$$(\Delta X_{\theta,\text{out}})^2 = \frac{1}{2}\left(|r|^2\epsilon + |t|^2\right) \tag{4.3.17}$$

The squeezing factor after the beamsplitter is

$$\epsilon + |t|^2(1-\epsilon) > \epsilon, \tag{4.3.18}$$

indicating that the squeezing is degraded. This turns out to be one of the most pernicious constraints towards using squeezed states for precision metrology. Any time we lose some of it, to anything, we are less able to exploit them for metrological gain! And loss is everywhere.

We have implemented attenuation using a beamsplitter. That said, it seems reasonable on physical grounds that absorptive loss could be seen in a similar way: by saying a portion of vacuum is mixed in. It turns out that a rigorous theory of absorptive loss does show that it can be treated by a beamsplitter approach, making this one of the most important models in quantum optics!

Before concluding, we also discuss the influence of a beamsplitter on photon number fluctuations. To evaluate it, we need to consider the second moment of the photon number at the output:

$$n_{\text{out}}^2 = b_1^\dagger b_1 b_1^\dagger b_1 = b_1^\dagger b_1^\dagger b_1 b_1 + b_1^\dagger b_1. \tag{4.3.19}$$

The expectation value of the quartic term is given by

$$\langle b_1^\dagger b_1^\dagger b_1 b_1 \rangle = |r|^4 \langle a_1^{\dagger 2} a_1^2 \rangle = |r|^4 \langle a_1^\dagger (a_1 a_1^\dagger - 1) a_1 \rangle. \tag{4.3.20}$$

Therefore

$$\langle b_1^\dagger b_1 b_1^\dagger b_1 \rangle = |r|^4 \langle n_{\text{in}}^2 \rangle + |t|^2 \langle n_{\text{in}} \rangle. \tag{4.3.21}$$

The variance then follows as

$$(\Delta n_{\text{out}})^2 = |r|^4 (\Delta n_{\text{in}})^2 + (|r|^2 - |r|^4) \langle n_{\text{in}} \rangle. \tag{4.3.22}$$

Noting that the fractional intensity output is $\ell = |r|^2$, we may write the result as

$$(\Delta n_{\text{out}})^2 = \ell^2 (\Delta n_{\text{in}})^2 + \ell(1-\ell) \langle n_{\text{in}} \rangle. \tag{4.3.23}$$

In the limit of large attenuation $\ell \to 0$, the variance becomes approximately $\ell \langle n_{\text{in}} \rangle = \langle n_{\text{out}} \rangle$ which corresponds to a Poissonian distribution of photons at the output.

This result (as well as the result for quadrature variances) depends critically on incorporating the second port. It is not entirely obvious from the derivation, but suppose we had assumed the mapping $b_1 = -r a_1$. In this case, the output commutator would be $[b_1, b_1^\dagger] = |r|^2 \neq 1$ for general $r$. Of course, this violates quantum mechanics (as it implies that we can have a product of quadrature variances below 1/2). But, taking this result, and doing the calculation *consistently*, one would find $\langle n_{\text{out}}^2 \rangle = |r|^4 \langle n_{\text{in}}^2 \rangle - |r|^4 \langle n_{\text{in}} \rangle + |r|^4 \langle n_{\text{in}} \rangle = |r|^4 \langle n_{\text{in}}^2 \rangle$. The last term arises from writing $b_1^\dagger b_1 b_1^\dagger b_1 = b_1^\dagger b_1^\dagger b_1 b_1 + |r|^2 b_1^\dagger b_1$ which comes from the use of the (incorrect!) commutator. The (incorrect) result which is obtained is what one would get if attenuation were *deterministic*. In other words, if loss mapped the photon number $n$ to $|r|^2 n$ in a deterministic fashion, then the variance of the photon number would indeed just scale by $|r|^4$. Of course, this is not how scattering works. There is a *probability* of scattering $|r|^2$ and this is enough to recover the correct result. Additionally, you see from the quantization of light into photons that a deterministic scattering map of the type $n \to |r|^2 n$ is inconsistent with photons appearing in integer amounts. The intuition that is often stated in quantum optics is that the beamsplitter feeds vacuum fluctuations in from the empty port in a way that preserves commutation relations. Indeed you see without the second port, the commutator would break (at which point we should have not calculated anything further) and we would violate the uncertainty principle. You are encouraged to convince yourself that the result we obtained for quadrature variances also would have not been obtained if we had take $b_1 = -r a_1$.

### 4.3.3 Homodyne detection

We now turn to the question of how one can experimentally access the statistical properties of a state of light, particularly in the quantum regime. For example, we may wish to characterize quantities such as the quadrature variances of a weak quantum state, such as a squeezed vacuum state. Unlike photon number, these observables are phase-sensitive, and therefore

require a measurement scheme capable of distinguishing between different quadratures (e.g., $X$ vs. $P$). However, most photodetectors naturally measure intensity, not phase, suggesting that some form of interferometric technique is required. In addition, states such as squeezed vacuum may contain very few photons, making direct detection challenging. It is therefore desirable to have a method that allows us to probe such weak states using measurements involving large numbers of photons.

A powerful approach that addresses these challenges is *homodyne detection*. The basic idea is to interfere the quantum state we wish to characterize (the signal) with a strong reference beam of known phase, known as the *local oscillator*. By mixing the two fields on a beamsplitter, the statistical properties of the signal are effectively transferred onto the strong beam, enabling detection with high signal-to-noise. Furthermore, by varying the phase of the local oscillator, one can control the relative phase between the two fields, allowing phase-sensitive measurements that probe different quadratures of the signal state.

We now introduce the basic optical element underlying homodyne detection: a 50:50 beamsplitter. We denote the two input modes by $a_1$ and $a_2$, and the two output modes by $b_1$ and $b_2$. The beamsplitter mixes the input fields linearly, and in the convention used here the transformation is

$$\begin{pmatrix} b_1 \\ b_2 \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} -1 & i \\ i & -1 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \end{pmatrix}. \tag{4.3.24}$$

Equivalently, the output operators are

$$b_1 = \frac{1}{\sqrt{2}} \left(-a_1 + i a_2\right), \tag{4.3.25}$$

$$b_2 = \frac{1}{\sqrt{2}} \left(i a_1 - a_2\right). \tag{4.3.26}$$

Since this transformation is unitary, it preserves the bosonic commutation relations. For example,

$$[b_i, b_j^\dagger] = \delta_{ij}. \tag{4.3.27}$$

The corresponding output photon number operators are

$$n_{b_1} = b_1^\dagger b_1, \qquad n_{b_2} = b_2^\dagger b_2. \tag{4.3.28}$$

Using the beamsplitter relations, these become

$$n_{b_1} = \frac{1}{2}(-a_1^\dagger - i a_2^\dagger)(-a_1 + i a_2) \tag{4.3.29}$$

$$= \frac{1}{2}\left(a_1^\dagger a_1 - i a_1^\dagger a_2 + i a_2^\dagger a_1 + a_2^\dagger a_2\right), \tag{4.3.30}$$

and

$$n_{b_2} = \frac{1}{2}(-i a_1^\dagger - a_2^\dagger)(i a_1 - a_2) \tag{4.3.31}$$

$$= \frac{1}{2}\left(a_1^\dagger a_1 + i a_1^\dagger a_2 - i a_2^\dagger a_1 + a_2^\dagger a_2\right). \tag{4.3.32}$$

It is especially useful to consider the sum and difference of the two output photocurrents. The sum is

$$n_{b_1} + n_{b_2} = a_1^\dagger a_1 + a_2^\dagger a_2, \tag{4.3.33}$$

so the total photon number is conserved by the beamsplitter.

The difference is

$$n_{b_1} - n_{b_2} = i\left(a_2^\dagger a_1 - a_1^\dagger a_2\right). \tag{4.3.34}$$

This operator will play a central role in homodyne detection. It shows that the difference signal at the two outputs is sensitive not simply to the intensity in one input mode alone, but to the interference between the two input fields.

At this stage we keep both input modes completely general. Later, one of these inputs will be taken to be a reference field (the local oscillator), and this will allow the output difference signal to probe specific field quadratures of the other mode.

We now assume that input mode $a_2$ is prepared in a coherent state and serves as the local oscillator:

$$a_2 \left|\alpha_{\rm LO}\right\rangle = \alpha_{\rm LO} \left|\alpha_{\rm LO}\right\rangle, \qquad \alpha_{\rm LO} = |\alpha_{\rm LO}| e^{i\theta}, \tag{4.3.35}$$

while $a_1$ is the signal mode and may be in an arbitrary quantum state.

From the previous section, the difference photocurrent operator is

$$I_- \equiv n_{b_1} - n_{b_2} = i\left(a_2^\dagger a_1 - a_1^\dagger a_2\right). \tag{4.3.36}$$

Taking the expectation value over the local-oscillator mode gives

$$\langle I_- \rangle = i\left\langle a_2^\dagger a_1 - a_1^\dagger a_2 \right\rangle \tag{4.3.37}$$

$$= i\left(\alpha_{\rm LO}^* \langle a_1 \rangle - \alpha_{\rm LO} \langle a_1^\dagger \rangle\right). \tag{4.3.38}$$

We define the signal quadrature in the standard form

$$X_\phi \equiv \frac{1}{\sqrt{2}}\left(a_1 e^{-i\phi} + a_1^\dagger e^{i\phi}\right). \tag{4.3.39}$$

Using $\alpha_{\rm LO} = |\alpha_{\rm LO}| e^{i\theta}$, we obtain

$$\langle I_- \rangle = i|\alpha_{\rm LO}| \left(e^{-i\theta}\langle a_1 \rangle - e^{i\theta}\langle a_1^\dagger \rangle\right) \tag{4.3.40}$$

$$= \sqrt{2}\,|\alpha_{\rm LO}|\,\langle X_{\theta+\pi/2} \rangle. \tag{4.3.41}$$

Thus, the mean difference current directly measures the quadrature of the signal mode shifted by $\pi/2$ relative to the local-oscillator phase in this beamsplitter convention. Equivalently, one may write the measured quadrature as $X_{\theta-\pi/2}$ after a redefinition of the local-oscillator phase origin.

It is also useful to write this at the operator level. Since the local oscillator is in a coherent state, we may write

$$a_2 = \alpha_{\rm LO} + \delta a, \tag{4.3.42}$$

where $\delta a$ describes the residual fluctuations of the local oscillator, with

$$\langle \delta a \rangle = 0, \qquad \langle \delta a^\dagger \delta a \rangle = 0, \qquad \langle \delta a \delta a^\dagger \rangle = 1. \tag{4.3.43}$$

Substituting into the difference current gives

$$I_- = i\left[(\alpha_{\rm LO}^* + \delta a^\dagger)a_1 - a_1^\dagger(\alpha_{\rm LO} + \delta a)\right] \tag{4.3.44}$$

$$= i\left(\alpha_{\rm LO}^* a_1 - \alpha_{\rm LO} a_1^\dagger\right) + i\left(\delta a^\dagger a_1 - a_1^\dagger \delta a\right) \tag{4.3.45}$$

$$= \sqrt{2}\,|\alpha_{\rm LO}|\, X_{\theta+\pi/2} + i\left(\delta a^\dagger a_1 - a_1^\dagger \delta a\right). \tag{4.3.46}$$

Therefore, in the limit of a strong local oscillator, $|\alpha_{\rm LO}| \gg 1$, the first term dominates and

$$I_- \approx \sqrt{2}\,|\alpha_{\rm LO}|\, X_{\theta+\pi/2}. \tag{4.3.47}$$

This is the basic principle of homodyne detection: by changing the phase $\theta$ of the local oscillator, one selects which signal quadrature is measured.

We now compute the mean and variance of the difference current more explicitly. The mean is

$$\langle I_- \rangle = \sqrt{2}\,|\alpha_{\rm LO}|\,\langle X_{\theta+\pi/2}\rangle. \tag{4.3.48}$$

For the variance, write

$$(\Delta I_-)^2 = \langle I_-^2 \rangle - \langle I_- \rangle^2. \tag{4.3.49}$$

Using

$$I_- = \sqrt{2}\,|\alpha_{\rm LO}|\, X_{\theta+\pi/2} + i\left(\delta a^\dagger a_1 - a_1^\dagger \delta a\right), \tag{4.3.50}$$

and the fact that the signal and local-oscillator modes are independent, the cross terms vanish upon taking expectation values. Therefore,

$$(\Delta I_-)^2 = 2|\alpha_{\rm LO}|^2 (\Delta X_{\theta+\pi/2})^2 + \left\langle \left[i(\delta a^\dagger a_1 - a_1^\dagger \delta a)\right]^2 \right\rangle. \tag{4.3.51}$$

The second term is

$$\left\langle \left[i(\delta a^\dagger a_1 - a_1^\dagger \delta a)\right]^2 \right\rangle = \langle a_1^\dagger a_1 \rangle \langle \delta a \delta a^\dagger \rangle \tag{4.3.52}$$

$$= \langle a_1^\dagger a_1 \rangle, \tag{4.3.53}$$

since all other terms vanish for a coherent-state fluctuation mode $\delta a$.

Thus the exact variance is

$$(\Delta I_-)^2 = 2|\alpha_{\rm LO}|^2 (\Delta X_{\theta+\pi/2})^2 + \langle a_1^\dagger a_1 \rangle. \tag{4.3.54}$$

In the strong-local-oscillator limit, the first term dominates, and one obtains

$$(\Delta I_-)^2 \approx 2|\alpha_{\rm LO}|^2 (\Delta X_{\theta+\pi/2})^2. \tag{4.3.55}$$

So both the mean and the variance of the difference photocurrent are directly proportional to the corresponding mean and variance of the selected signal quadrature. This is why homodyne detection provides a direct experimental probe of quadrature statistics, including squeezing.

### 4.3.4 Sub shot-noise interferometry

As we know from elementary courses on waves, interferometry gives us a tool to measure small changes in length by translating them to phase shifts that manifest as changes in the intensity of light. We will perform a quantum analysis of an interferometer and show that the resolvable phase shift is limited by vacuum fluctuations. Then we show that by using squeezed states, the sensitivity can be enhanced.

Let us consider a Mach-Zehnder interferometer. Light enters one port of a 50/50 beamsplitter with the other port being empty. The two arms undergo a relative phase shift $\phi$ and are then recombined by another 50/50 beamsplitter. As we know quantum mechanically, the empty port corresponds to vacuum being sent in, and we must keep track of this. Let the two input arms have lowering operators $a_1, a_2$. After passing through the beamsplitter, they are mapped to

$$b_1 = \frac{1}{\sqrt{2}}\left(-a_1 + ia_2\right), b_2 = \frac{1}{\sqrt{2}}\left(ia_1 - a_2\right). \tag{4.3.56}$$

Let us assume it is the second arm that experiences an additional phase shift, so that the operators just before the second 50/50 beamsplitter may be written as

$$c_1 = \frac{1}{\sqrt{2}}\left(-a_1 + ia_2\right), c_2 = \frac{1}{\sqrt{2}}\left(ia_1 - a_2\right)e^{-i\theta}. \tag{4.3.57}$$

At the second 50/50 beamsplitter, the output operators in the first output port may be written as

$$d_1 = \frac{1}{\sqrt{2}}\left(-c_1 + ic_2\right) = \frac{1}{\sqrt{2}}\left(-\frac{1}{\sqrt{2}}\left(-a_1 + ia_2\right) + i\frac{1}{\sqrt{2}}\left(ia_1 - a_2\right)e^{-i\theta}\right), \tag{4.3.58}$$

which may be simplified as

$$d_1 = \frac{1}{2}\left(a_1(1 - e^{-i\theta}) - ia_2(1 + e^{-i\theta})\right). \tag{4.3.59}$$

The second output port operator may be written as

$$d_2 = \frac{1}{\sqrt{2}}\left(ic_1 - c_2\right) = \frac{1}{\sqrt{2}}\left(i\frac{1}{\sqrt{2}}\left(-a_1 + ia_2\right) - \frac{1}{\sqrt{2}}\left(ia_1 - a_2\right)e^{-i\theta}\right), \tag{4.3.60}$$

which may be similarly simplified as

$$d_2 = \frac{1}{2}\left(-ia_1(1 + e^{-i\theta}) + a_2(e^{-i\theta} - 1)\right). \tag{4.3.61}$$

It is helpful to express the overall transformation in terms of an $S$ matrix, which is found to be:

$$S = \frac{1}{2}\begin{pmatrix} (1 - e^{-i\theta}) & -i(1 + e^{-i\theta}) \\ -i(1 + e^{-i\theta}) & (e^{-i\theta} - 1) \end{pmatrix} = ie^{-\frac{i\theta}{2}}\begin{pmatrix} \sin\frac{\theta}{2} & -\cos\frac{\theta}{2} \\ -\cos\frac{\theta}{2} & -\sin\frac{\theta}{2} \end{pmatrix}, \tag{4.3.62}$$

It is easy to see from the trigonometric form that this transformation is unitary.

The photon numbers associated with the output ports may be written as

$$d_1^\dagger d_1 = \sin^2\left(\frac{\theta}{2}\right)a_1^\dagger a_1 + \cos^2\left(\frac{\theta}{2}\right)a_2^\dagger a_2 - \sin\left(\frac{\theta}{2}\right)\cos\left(\frac{\theta}{2}\right)\left(a_1^\dagger a_2 + a_2^\dagger a_1\right) \tag{4.3.63}$$

and

$$d_2^\dagger d_2 = \cos^2\left(\frac{\theta}{2}\right) a_1^\dagger a_1 + \sin^2\left(\frac{\theta}{2}\right) a_2^\dagger a_2 + \sin\left(\frac{\theta}{2}\right)\cos\left(\frac{\theta}{2}\right)(a_1^\dagger a_2 + a_2^\dagger a_1). \tag{4.3.64}$$

Let us now consider the configuration of this interferometer a bit more carefully now. In practice, the phase shift we are trying to measure is small. We would like our signal to be zero when there is no phase shift and to increase from zero when the source of the phase shift is present. This leads to two choices in operating this interferometer. The first is that we will "bias" the interferometer so that in the absence of our source phase shift, there is a phase-shift between the arms of $\pi/2$, so the overall phase shift is $\pi/2 + \theta$. Thus we make the mapping $\theta \to \frac{\pi}{2} + \theta$ in the above expressions. When the phase shift is exactly $\pi/2$ ($\theta = 0$) then we see from the above photon-number expressions that classically, when the $a_2$ port is empty, the photon number in each arm is identical. Since we'd like the signal to increase from zero, we subtract the two photon numbers. Therefore, we define our signal to be

$$S = d_1^\dagger d_1 - d_2^\dagger d_2. \tag{4.3.65}$$

Since $\theta$ is small, we will expand around $\theta = 0$, in which case the signal is given by (to linear order in $\theta$)

$$S = \theta(a_1^\dagger a_1 - a_2^\dagger a_2) - (a_1^\dagger a_2 + a_2^\dagger a_1) \equiv \theta(a_1^\dagger a_1 - a_2^\dagger a_2) + C. \tag{4.3.66}$$

The mean value of the signal in the empty port configuration is

$$\langle S\rangle = \theta\langle a_1^\dagger a_1\rangle. \tag{4.3.67}$$

The variance of the signal can be found quickly as follows. The expectation value of $S^2$ has a term which is quadratic in $\theta$, a term which is $C^2$, and a cross term. The cross term makes no contribution because the expectation values of operators linear and cubic in $a_2$ vanish. The $\theta^2$ term has a variance of $\theta^2\Delta(a_1^\dagger a_1)^2$ which we will find is small[17]. Taking the input field to be a coherent state $|\alpha\rangle$ with $\alpha$ real, $X$ may be approximated via

$$C \approx -\alpha(a_2 + a_2^\dagger) = -\sqrt{2}\alpha X_0, \tag{4.3.68}$$

where $X_0$ is a quadrature operator corresponding to $a_2$. Therefore, the total variance may be written as

$$(\Delta S)^2 = \theta^2\alpha^2 + 2\alpha^2(\Delta X_0)^2, \tag{4.3.69}$$

which for the vacuum state may be written as (using $(\Delta X_0)^2 = 1/2$)

$$(\Delta S)^2 = (1+\theta^2)\alpha^2 \approx \alpha^2. \tag{4.3.70}$$

The variance comes entirely from vacuum fluctuations of the empty port, amplified by the mean signal! The measure of the signal-to-noise ratio of the measurement is given by the relative variance

$$\frac{\Delta S}{\langle S\rangle} = \frac{1}{\theta}\frac{1}{\sqrt{\langle n\rangle}}. \tag{4.3.71}$$

[17]This follows from the two input ports having no correlations, in which case the variance of the difference of photon numbers is simply the sum of the variances plus twice the covariance.

This tells us that the weaker the phase shift, the harder it is to see, but that by having many photons, we can resolve it more easily. You can think of each photon as sampling the phase corresponding to $n$ trials of an experiment giving the expected $\sqrt{n}$ behavior. This $1/\sqrt{n}$ scaling is called the shot-noise limit and is the variance imposed by simply having the empty port present.

The variance can be reduced by sending a squeezed vacuum state into the empty port. We shall assume that $\sinh^2 r \ll |\alpha|^2$: that the number of photons in the squeezed vacuum field is far less than the number of photons in the coherent laser field (this in practice is more than well-satisfied). Repeating the calculation for squeezed input under these approximations, the mean signal is unchanged. The variance however becomes $2\alpha^2(\Delta X_0)^2$ (neglecting $\theta^2$). Assuming that our squeezed vacuum is squeezed in the right quadrature, the variance becomes $\alpha^2 e^{-2r}$ and our relative variance becomes

$$\frac{\Delta S}{\langle S\rangle} = \frac{1}{\theta}\frac{e^{-r}}{\sqrt{\langle n\rangle}}, \tag{4.3.72}$$

corresponding to an enhancement in the signal-to-noise ratio for the same intensity. This is a defining property of squeezed states and is what is currently used at advanced LIGO to detect gravitational waves. Gravitational waves correspond to propagating distortions of the spacetime metric which lead to changes in the distances between points in space. These changes can be measured interferometrically by detecting the corresponding phase shift of light. The main problem however is that these phase shifts are extraordinarily small. Therefore even with high intensity inputs into the interferometer, the signal-to-noise ratio is insufficient to resolve many different gravitational wave events. While one can always crank up the intensity to get a better signal-to-noise ratio, in practice the interferometers have a maximum laser intensity that they can handle. The injection of squeezed vacuum into the empty port gives lower variance for the same intensity, mimicking an intensity enhanced by $e^r$.

The principle of using squeezed vacuum can be used for any system which is limited by the shot noise of light and these ideas are now being explored in microscopy, biological imaging, and magnetometry, among other fields.

# Chapter 5

# Quantum description of nonlinear optics

In this unit, we will use the apparatus developed in this unit describe how nonlinear media lead to the generation of quantum mechanical states of light such as entangled photons and squeezed light. And we will conclude by illustrating how quantum light states allow measurement sensitivities which can exceed what is possible classically. In this section, we will make use of the more general theory of quantized electromagnetic fields in nonlinear media.

## 5.1 Heisenberg description of quantum nonlinear optics

As a first example, we will consider the process of parametric down conversion treated from a quantum mechanical perspective. Relative to the classical treatment we pursued in Unit 2, the main new effects will be that (a) the presence of a pump can lead to the generation of photon pairs[1], some of which display quantum entanglement, and (b) squeezed states of light can be generated. The single-mode squeezed states that we studied in Unit 4 may be generated using degenerate parametric down conversion.

In what follows, we will consider a physical situation which is very similar to that which we considered when analyzing parametric down-conversion / difference-frequency generation. We consider a $\chi^{(2)}$ nonlinear crystal of length $L$ along some direction $z$ and we illuminate this crystal with a strong pump wave which propagates along the $z$ direction. Unlike the case of difference-frequency generation, we will not explicitly illuminate the crystal with any lower-frequency fields (at frequency $\omega_1$) that could combine with the pump to generate a difference frequency (at frequency $\omega_2 = \omega_3 - \omega_1$. Classically, if there are no $\omega_1$ or $\omega_2$ fields then nothing happens. Quantum mechanically, zero-point fluctuations of the field provide a fluctuating seed that can enable difference-frequency generation, manifesting as pairs of photons.

The starting point for the quantum analysis is the Heisenberg equations of motion. In Unit 4, we showed that these were "simply" operator versions of the Maxwell equations. In what follows, it will be helpful to track parts of the field with positive versus negative frequency (as

[1]We stated this was the photon picture of parametric down conversion in Unit 2, but could not prove this, of course.

was also the case classically). To do this, we introduce the so-called positive and negative frequency parts of the electric field operator:

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}^{(+)}(\mathbf{r},t) + \mathbf{E}^{(-)}(\mathbf{r},t). \tag{5.1.1}$$

The positive and negative frequency parts of the field operator are defined such that

$$\mathbf{E}^{(\pm)}(\mathbf{r},t) = \int_0^\infty \frac{d\omega}{2\pi} e^{\mp i\omega t} E^{(\pm)}(\mathbf{r},\omega). \tag{5.1.2}$$

Let us now consider for example the Fourier component of the electric field operator at $\omega$ (associated with $e^{-i\omega t}$). It follows from these definitions, and the operator Maxwell equations, that:

$$\nabla\times\nabla\times\mathbf{E}^{(+)}(\mathbf{r},\omega) - \epsilon(\omega)\frac{\omega^2}{c^2}\mathbf{E}^{(+)}(\mathbf{r},\omega) = \mu_0\omega^2\mathbf{P}^{(+)}(\mathbf{r},\omega), \tag{5.1.3}$$

where the positive frequency part of the polarization is defined similarly to the electric field. This equation is complicated by the fact that $\epsilon(\omega)$ a tensor and so the double curl is not necessarily equal to the negative Laplacian. As we did in Unit 2, we assume weak anisotropy, which allows us to write

$$-\left(\nabla^2 + \epsilon(\omega)\frac{\omega^2}{c^2}\right)\mathbf{E}^{(+)}(\mathbf{r},\omega) = \mu_0\omega^2\mathbf{P}^{(+)}(\mathbf{r},\omega). \tag{5.1.4}$$

Physically, we know that phase-matching has a strong impact on which frequencies and wavevectors of light get efficiently generated, and we have mentioned previously that this phase-matching manifests in the quantum mechanical theory as energy-momentum conservation. We thus expect that there is in general, for monochromatic plane-wave pumping, a narrow range of frequencies that is efficiently generated by this parametric process. This allows us to write

$$\mathbf{E}^{(+)}(\mathbf{r},\omega) = \mathbf{E}_1^{(+)}(\mathbf{r},\omega) + \mathbf{E}_2^{(+)}(\mathbf{r},\omega), \tag{5.1.5}$$

where $E_{1,2}$ are operators with significant spectral weight only around special frequencies $\omega_{1,2}$ associated with phase-matching. We will assume that the bandwidths $\Delta\omega_{1,2} \ll \omega_{1,2}$ which is often a reasonable approximation. Note that the assumption of In Unit 2, we mostly concerned ourselves with the collinear case where the generated frequencies were parallel to the pump wavevector. In the quantum case, we will consider the more general case in which the generated fields are not parallel to the pump wavevector. This turns out to be important for the generation of entangled photons. Therefore, we shall further Fourier transform the field operators in space via

$$\mathbf{E}^{(+)}(\mathbf{k}_\perp,z,\omega) = \mathbf{E}_1^{(+)}(\mathbf{k}_\perp,z,\omega) + \mathbf{E}_2^{(+)}(\mathbf{k}_\perp,z,\omega). \tag{5.1.6}$$

For each $\mathbf{k}_\perp$, since the spread in frequencies is small, we expect the spread in longitudinal wavenumbers to be small as well, and so the overall spread of wavevectors is small. Therefore, we expect there to be a relatively well-defined polarization vector. We therefore separate it from the field operators as

$$E^{(+)}(\mathbf{k}_\perp,z,\omega) = E_1^{(+)}(\mathbf{k}_\perp,z,\omega)\hat{\epsilon}_{1,\mathbf{k}} + E_2^{(+)}(\mathbf{k}_\perp,z,\omega)\hat{\epsilon}_{2,\mathbf{k}}, \tag{5.1.7}$$

where $\mathbf{k} = (\mathbf{k}_\perp, k_z)$. We can use this decomposition that we have worked out to write equations of motion for $E_{1,2}$ as follows. For $E_1$, we have

$$-\left(\frac{d^2}{dz^2} + \left(\frac{n_{\text{eff}}(\omega)\omega}{c}\right)^2 - k_\perp^2\right) E_1^{(+)}(\mathbf{k}_\perp, z, \omega) = \mu_0\omega^2 \hat{\epsilon}_{1,\mathbf{k}} \cdot \mathbf{P}_1^{(+)}(\mathbf{k}_\perp, z, \omega), \tag{5.1.8}$$

where $n_{\text{eff}}^2 \equiv \hat{\epsilon}_{1,\mathbf{k}} \cdot \epsilon(\omega) \cdot \hat{\epsilon}_{1,\mathbf{k}}$, where I have taken real polarizations. We define $k_{1,z}(\omega) \equiv \sqrt{\left(\frac{n_{\text{eff}}(\omega)\omega}{c}\right)^2 - k_\perp^2}$ and write

$$-\left(\frac{d^2}{dz^2} + k_{1,z}^2(\omega)\right) E_1^{(+)}(\mathbf{k}_\perp, z, \omega) = \mu_0\omega^2 \hat{\epsilon}_{1,\mathbf{k}} \cdot \mathbf{P}_1^{(+)}(\mathbf{k}_\perp, z, \omega), \tag{5.1.9}$$

We now make a slowly varying envelope approximation. We write

$$E_1^{(+)}(\mathbf{k}_\perp, z, \omega) = A_1^{(+)}(\mathbf{k}_\perp, z, \omega) e^{ik_{1,z}(\omega_1)z}, \tag{5.1.10}$$

and neglect terms associated with the second derivative of the slow operator. This turns the left-hand side into

$$-\left(2ik_{1,z}(\omega_1)\partial_z A_1^{(+)}(\mathbf{k}_\perp, z, \omega) + \left(k_{1,z}^2(\omega) - k_{1,z}^2(\omega_1)\right) A_1^{(+)}(\mathbf{k}_\perp, z, \omega)\right). \tag{5.1.11}$$

This of course is the same type of structure we ran into when deriving pulse propagation equations. We may now expand $k_{1,z}(\omega)$ around $\omega_1$, writing

$$k_{1,z}(\omega) = k_{1,z}(\omega_1) + \sum_{m=1}^{\infty} \frac{\beta_m}{m!}(\omega - \omega_1)^m. \tag{5.1.12}$$

We may therefore write an envelope equation for a time-domain field $\tilde{A}(\mathbf{k}_\perp, z, t)$ by taking $(\omega - \omega_1) \to i\partial_t$. The left-hand side of such an equation is

$$-\left(2ik_{1,z}(\omega_1)\partial_z \tilde{A}_1^{(+)}(\mathbf{k}_\perp, z, t) + 2k_{1,z}(\omega_1) \sum_{m=1}^{\infty} \frac{i^m \beta_m}{m!} \partial_t^m \tilde{A}_1^{(+)}(\mathbf{k}_\perp, z, t)\right). \tag{5.1.13}$$

The polarization terms in the time-domain may be written as (in envelope form)

$$\mu_0\omega_1^2 \hat{\epsilon}_{1,\mathbf{k}} \cdot \tilde{\mathbf{P}}_1^{(+)}(\mathbf{k}_\perp, z, t), \tag{5.1.14}$$

and so putting everything together, we have

$$\partial_z \tilde{A}_1^{(+)}(\mathbf{k}_\perp, z, t) = \sum_{m=1}^{\infty} \frac{i^{m+1}\beta_m}{m!} \partial_t^m \tilde{A}_1^{(+)}(\mathbf{k}_\perp, z, t) + \frac{i\omega_1^2}{2\epsilon_0 c^2 k_{1,z}(\omega_1)} \hat{\epsilon}_{1,\mathbf{k}} \cdot \tilde{\mathbf{P}}_1^{(+)}(\mathbf{k}_\perp, z, t). \tag{5.1.15}$$

This is a quantum mechanical version of the pulse-propagation equation in a nonlinear medium. This is somewhat more general that we need for our analysis, but a similar equation will come up later, and essentially follows immediately from the techniques we have developed throughout the course. You may notice that this equation formally is identical to the classical

equation, just with operators. That will be a theme of our explorations of quantum nonlinear optics, and we will often exploit this to simply "write" a quantized version of an equation based on the corresponding classical equation. This works particularly when operator ordering ambiguity is not present (which occurs when we have linear equations in a field and its conjugate). Despite this, it is in your instructor's view remarkable that replacing the classical field amplitudes by operators for these classically derived equations is consistent with the canonical commutation relations!

## 5.2 Parametric down-conversion in a bulk crystal

Let us now analyze a relatively simple case of parametric down-conversion in which the pump is a monochromatic plane-wave propagating along the $z$-direction, and so dispersion is negligible. This case, which maps to the classical case we considered in Unit 2, allows us to drop the time-variable, treating the fields as being in a steady-state at the exit of the crystal[2]. We are left with

$$\partial_z \tilde{A}_1^{(+)}(\mathbf{k}_{1,\perp}, z, t) = \frac{i\omega_1^2}{2\epsilon_0 c^2 k_{1,z}(\omega_1)} \hat{\epsilon}_{1,\mathbf{k}} \cdot \tilde{\mathbf{P}}_1^{(+)}(\mathbf{k}_{1,\perp}, z, t), \tag{5.2.1}$$

where I have taken $\mathbf{k}_\perp \to \mathbf{k}_{1,\perp}$ in order to remind us that we're taking about the $\omega_1$ field.

We now consider the polarization. In order to proceed, we note that the pump field has many photons in it, and that it is negligibly depleted by parametric generation. In this case, it is clear that the quantum fluctuations of this field are not important: it is the strong average field which generates the photon pairs, not the weak fluctuations. Therefore, we write our electric field operator as[3]

$$\mathbf{E}^{(+)}(\mathbf{r}, t) \to \left( A_3 e^{i\mathbf{k}_3 \cdot \mathbf{r} - i\omega_3 t} \hat{\epsilon}_3 + \text{c.c.} \right) + \mathbf{E}^{(+)}(\mathbf{r}, t), \tag{5.2.2}$$

where I have defined the wavevector and frequency of the pump to be $\mathbf{k}_3, \omega_3$ in keeping with the notation from our classical treatment. The term $\mathbf{E}_Q^{(+)}(\mathbf{r}, t)$ represents the "quantum" part of the field operator, and should be understood as representing everything but the strong classical oscillation. Therefore the number of photons per unit time associated with this field is much smaller than that associated with the classical pump part. The polarization can be written as

$$P_i(\mathbf{r}, t) = \epsilon_0 \chi_{ijk}^{(2)} E_i(\mathbf{r}, t) E_j(\mathbf{r}, t). \tag{5.2.3}$$

Clearly, the leading-order operator behavior comes from terms linear in the strong classical pump. Since we are looking at the equation of motion for $A_1^{(+)}$ which corresponds to an $e^{-i\omega_1 t}$ oscillation, it is clear that the only way to get the frequencies on the left and right-hand sides to work out is if we take $A_3 e^{i\mathbf{k}_3 \cdot \mathbf{r} - i\omega_3 t} \hat{\epsilon}_3$ from the classical field and consider only the negative frequency part of the quantized field near frequency $\omega_2$. Defining

$$\mathbf{E}^{(-)}(\mathbf{r}, t) = \int \frac{d^2 k_\perp}{(2\pi)^2} \frac{d\omega}{2\pi} \mathbf{E}^{(-)}(\mathbf{k}_\perp, z, \omega) e^{-i\mathbf{k}_\perp \cdot \rho + i\omega t}, \tag{5.2.4}$$

[2] If this sits uncomfortably with you, note that one can also get the same equation by going into the co-moving frame at the group velocity of the field and dropping second- and higher-order dispersion.

[3] Technically, this constitutes a redefinition of the field operator. It is analogous to writing $a \to a + \alpha$ when transforming with the displacement operator.

and defining envelopes an analogous way, it is straightforward to show

$$\partial_z \tilde{A}_1^{(+)}(\mathbf{k}_{1,\perp}, z, \omega) = \frac{2i\omega_1^2 d_{\text{eff}} A_3}{c^2 k_{1,z}(\omega_1)} \tilde{A}_2^{(-)}(\mathbf{k}_{3,\perp} - \mathbf{k}_{1,\perp}, z, \omega_3 - \omega) e^{i\Delta k_z z}, \tag{5.2.5}$$

with $\Delta k_z = k_{3,z} - k_{2,z}(\omega_2) - k_{1,z}(\omega_1)$. This equation is more or less identical to the one we encountered for difference frequency generation, just with operators. But the equation is linear and so its solution is the same. We may immediately write that for $z = L$ we have

$$\begin{aligned} A_1^{(+)}(\mathbf{k}_{1,\perp}, L, \omega) = &\left(\cosh(\lambda L) - \frac{i\Delta k_z}{2\lambda}\sinh(\lambda L)\right) A_1^{(+)}(\mathbf{k}_{1,\perp}, 0, \omega) \\ &+ \frac{\kappa_1}{\lambda}\sinh(\lambda L) A_2^{(-)}(\mathbf{k}_{2,\perp}, 0, \omega_3 - \omega), \end{aligned} \tag{5.2.6}$$

where $\lambda^2 = \frac{4\omega_1^2\omega_2^2 d_{\text{eff}}^2}{k_{1,z}k_{2,z}c^4}|A_3|^2 - \frac{(\Delta k_z)^2}{4} = \kappa_1\kappa_2^* - \frac{(\Delta k_z)^2}{4}$ and $\kappa_i = \frac{2id_{\text{eff}}\omega_i^2 A_3}{c^2 k_{i,z}(\omega_i)}$. This is a so-called two-mode Bogoliubov transformation. It is of the form $a \to \mu a + \nu b^\dagger$. For the transformation to be valid, it must preserve the commutation relations at [4]. Thus, we must check

$$1 = \left|\left(\cosh(\lambda L) - \frac{i\Delta k_z}{2\lambda}\sinh(\lambda L)\right)\right|^2 - \left|\frac{\kappa_1}{\lambda}\right|^2 \sinh^2(\lambda L) R_C, \tag{5.2.7}$$

where $R_C = \frac{[A_2^{(+)}(\mathbf{k}_{2,\perp},0,\omega_3-\omega), A_2^{(-)}(\mathbf{k}_{2,\perp},0,\omega_3-\omega)]}{[A_1^{(+)}(\mathbf{k}_{1,\perp},0,\omega), A_1^{(-)}(\mathbf{k}_{1,\perp},0,\omega)]}$ is a ratio of commutators.

To proceed, we need to evaluate this ratio. It is equal to[5]

$$R_C = \frac{n_1 \cos\theta_1 \omega_2}{n_2 \cos\theta_2 \omega_1}. \tag{5.2.8}$$

With this value, it is straightforward to show that the commutation relations are preserved. With this derivation, we can convert Eq. II.6 into a more standard form involving creation and annihilation operators as

$$a_1(\mathbf{k}_{1,\perp}, L, \omega_1) = \mu(\mathbf{k}_{1,\perp}, \omega_1) a_1(\mathbf{k}_{1,\perp}, 0, \omega_1) + \nu(\mathbf{k}_{1,\perp}, \omega_1) a_2^\dagger(\mathbf{k}_{2,\perp}, 0, \omega_3 - \omega_1), \tag{5.2.9}$$

with

$$\mu(\mathbf{k}_{1,\perp}, \omega_1) = \cosh(\lambda L) - \frac{i\Delta k_z}{2\lambda}\sinh(\lambda L), \nu(\mathbf{k}_{1,\perp}, \omega_1) = i\sqrt{\frac{\kappa_1\kappa_2^*}{\lambda^2}}\sinh(\lambda L). \tag{5.2.10}$$

where these operators are normalized such that $[a(\mathbf{k}, \omega), a^\dagger(\mathbf{k}', \omega')] = (2\pi)^3\delta(\mathbf{k} - \mathbf{k}')\delta(\omega - \omega')$. The coefficients can be expressed in terms of $\mathbf{k}_{\perp,1}, \omega_1$ alone since the other wavevector and frequency are determined. Further $k_z$ is determined by $\mathbf{k}_\perp$ and $\omega$. It is somewhat inconvenient to work with delta-function normalized operators since we are accustomed to working with discretized operators. This however can be done straightforwardly by a box quantization type argument. Defining $a_{\mathbf{k}_\perp,\omega} \equiv \mathcal{N} a(\mathbf{k}_\perp, \omega)$ such that $[a_{\mathbf{k}_\perp,\omega}, a^\dagger_{\mathbf{k}_\perp,\omega}] = 1$ requires $\mathcal{N}^2 = \frac{2\pi A}{\Delta\omega}$ where $\Delta\omega$ is a small frequency separation between different allowed frequencies of light (technically this is zero) and $A$ is the area of a large box[6].

[4] We should see the evolution from $z = 0$ to $z = L$ as a Heisenberg transformation of operators. For it to be valid, the transformation must be unitary, and any commutators proportional to the identity should be preserved under the transformation.

[5] To see this, Fourier transform the box quantized expression for the field operator.

[6] You see why the delta function approach can be nice: no need for the large box!

### 5.2.1 Spontaneous parametric fluorescence

Using the discrete version of the operators, it is relatively easy to do two things now. One, let us calculate how many photons are generated in parametric down conversion. It follows immediately from our Bogoliubov transformation that the number of photons in "mode 1" (defined to be the photons near frequency $\omega_1$) is given by

$$n_1 = \sum_{\mathbf{k}_{1,\perp},\omega_1} \frac{\kappa_1 \kappa_2^*}{\lambda^2} \sinh^2(\lambda L). \tag{5.2.11}$$

Since $\mathbf{k}_\perp, \omega$ are effectively continuous, we may write them as integrals as follows

$$n_1 = AT \int \frac{d^2k_{1,\perp}}{(2\pi)^2} \frac{d\omega}{(2\pi)} \frac{\kappa_1 \kappa_2^*}{\lambda^2} \sinh^2(\lambda L), \tag{5.2.12}$$

where $T$ is a quantization time (a time box, if you will) that sets $\Delta\omega = 2\pi/T$. We may interpret $n_1/T$ as a rate of photons being generated, $R$. It is useful to also spectrally resolve this rate, as we can always use a spectrometer to resolve these parametric photons by frequency. Thus we write

$$\frac{dR}{d\omega} = A \int \frac{d^2k_{1,\perp}}{(2\pi)^3} \frac{\kappa_1 \kappa_2^*}{\lambda^2} \sinh^2(\lambda L). \tag{5.2.13}$$

The quantity multiplying the area is proportional to intensity: $\frac{dR}{d\omega} = \frac{A}{\hbar\omega_1}\frac{dI_1}{d\omega}$ per unit frequency (and is an intensive quantity), which we write as

$$\frac{1}{\hbar\omega_1}\frac{dI_1}{d\omega} = \int \frac{d^2k_{1,\perp}}{(2\pi)^3} \frac{4\omega_1^2\omega_2^2 d_{\text{eff}}^2 |A_3|^2 L^2}{k_{1,z}(\mathbf{k}_{1,\perp},\omega_1)k_{2,z}(\mathbf{k}_{2,\perp},\omega_2)c^4} \text{sinhc}^2\left(\sqrt{\frac{4\omega_1^2\omega_2^2 d_{\text{eff}}|A_3|^2L^2}{k_{1,z}k_{2,z}c^4} - \frac{(\Delta k_z)^2}{4}}L\right), \tag{5.2.14}$$

where I have defined $\text{sinhc}\ x = \sinh x/x$.

The number of photons in the second set of modes can be found to be equal to $n_1$ (replacing $1 \leftrightarrow 2$ in the Bogoliubov transformation does not change the coefficient of the creation operator part) which is expected based on the photon-pair picture of parametric down-conversion. This also automatically implies that the Manley-Rowe relation we encountered in Unit 2 ($\frac{I_2}{\omega_2} - \frac{I_1}{\omega_1} = 0$) is satisfied. While this is all suggestive of the two-photon picture, we would like to show more directly that the wavefunction has a sum of photon pairs. To do this, we note that the transformation $a \to \mu a + \nu b^\dagger$ corresponds to a unitary

$$U = \exp\left[\zeta a^\dagger b^\dagger - \zeta^* ab\right], \tag{5.2.15}$$

where $\zeta = re^{i\phi}, \mu = \cosh r, \nu = e^{i\phi}\sinh r$. There is one such unitary per pair of photon modes generated, and so we can write the wavefunction after parametric generation as

$$|\psi\rangle = \bigotimes_{\mathbf{k}_\perp,\omega} \exp\left[\zeta_{\mathbf{k}_\perp,\omega} a^\dagger_{\mathbf{k}_\perp,\omega} b^\dagger_{-\mathbf{k}_\perp,(\omega_3-\omega)} - \zeta^*_{\mathbf{k}_\perp,\omega} a_{\mathbf{k}_\perp,\omega} b_{-\mathbf{k}_\perp,(\omega_3-\omega)}\right]|0\rangle. \tag{5.2.16}$$

If the nonlinearity is weak such that the expected number of photons is much less than 1, then we may approximate this as

$$|\psi\rangle \approx |0\rangle + \sum_{\mathbf{k}_\perp,\omega} \zeta_{\mathbf{k}_\perp,\omega} |1_{\mathbf{k}_\perp,\omega} 1_{-\mathbf{k}_\perp,(\omega_3-\omega)}\rangle. \tag{5.2.17}$$

This shows that the wavefunction is a superposition of vacuum and a bunch of photon pairs whose amplitude depend on the phase-matching condition and satisfy exact transverse momentum and energy conservation. The peak pair amplitude occurs when phase-matching is satisfied. This calculation therefore establishes the photon picture of parametric generation that we have stated in our classical treatment of the subject. We conclude this section by noting that this effect is usually called parametric fluorescence and corresponds to a low flux of photon pairs emanating from the system[7].

**Generation of entangled photons**

The photons emitted in spontaneous parametric down-conversion have quantum correlations known as entanglement. Entanglement occurs when a wavefunction cannot be separated into a product of wavefunctions $|\psi_{1,2}\rangle \neq |\phi_1\rangle|\phi_2\rangle$ for some single-channel states $\phi_1, \phi_2$. The correlation structure is tied into the physics of phase-matching in a direct and beautiful way that we will explore here. Let us consider the limit in which nonlinearity is weak and we do not generate too many pairs at once. We will also focus on the case of degenerate parametric down-conversion. In that case we have the intensity of a pair of photons at $\mathbf{k}_1 s_1, \mathbf{k}_2 s_2$ where $s_{1,2} = o, e$ denotes the polarization state, is directly proportional to

$$\mathrm{sinc}^2\left(\frac{1}{2}(\Delta k_z)L\right), \tag{5.2.18}$$

where

$$\Delta k_z = k_{p,z} - k_{1,z} - k_{2,z}, \tag{5.2.19}$$

with

$$k_{i,z} = \sqrt{n^2_{s_i}(\omega_i, \theta)\frac{\omega_i^2}{c^2} - \mathbf{k}_\perp^2}, \tag{5.2.20}$$

where $s_i$ is the polarization state of photon $i$. For a given c-axis orientation of a crystal ($\beta$-BBO is one of the most common for SPDC), one can plot the in-plane wavevectors $k_x, k_y$ which lead to phase matching. The structure of this zero-mismatch contour for Type-I and Type-II phase matching is quite different. In the Type-I case for BBO (which is negative uniaxial) $e \to o, o$, the contour is a circle centered around $k_x, k_y = 0$ and so our wavefunction is a superposition of pairs of the form

$$a^\dagger_{\mathbf{k}_\perp, o} a^\dagger_{-\mathbf{k}_\perp, o}|0\rangle = |\mathbf{k}o, -\mathbf{k}o\rangle. \tag{5.2.21}$$

This state is separable and does not have entanglement (although once we consider the possibility of non-degenerate down-conversion there is spectral entanglement). In the type-II case however ($e \to e + o$), the contour of angles for the extraordinary photon and ordinary photon are different due to the asymmetry of positive and negative angle with respect to the $z$ axis (for general $c$ axis orientations). We can think of there being one cone of angles corresponding to extraordinary polarized photons and one cone corresponding to ordinary polarized photons. They intersect at two wavevectors $\mathbf{k}, \mathbf{k}$ Where they intersect, there is

[7] In principle it is possible to have many pairs emanate if the pump is sufficiently strong: this generates something called bright squeezed vacuum, not to be confused with displaced squeezed vacuum.

polarization entanglement. In particular, we see that there is a contribution to the overall photon state of the form

$$|\psi\rangle \sim a^\dagger_{\mathbf{k}_\perp,e} a^\dagger_{-\mathbf{k}_\perp,o}|0\rangle + a^\dagger_{\mathbf{k}_\perp,o} a^\dagger_{-\mathbf{k}_\perp,e}|0\rangle = |\mathbf{k}e, -\mathbf{k}o\rangle + |\mathbf{k}o, -\mathbf{k}e\rangle, \tag{5.2.22}$$

which corresponds to a so-called polarization-entangled state. It has the following property. Let's say I spatially filter out the two paths corresponding to $\mathbf{k}, -\mathbf{k}$ in the far-field. Then if I measure the photon at only $\mathbf{k}$, it has a 50% probability of being either $e$ or $o$ polarized. Meanwhile, if I measure the photon at $\mathbf{k}$ and find $e$ polarization, the other photon in the $-\mathbf{k}$ path is always $o$ polarized. These types of correlations are non-classical and have proven useful for a variety of applications from quantum key distribution to quantum computation.

**Heralded photons**

Although the type-I case seems less useful, it can be used as a source for so-called *heralded* single photons. Let us imagine the following thought experiment. Suppose that we have the state $\mathbf{k}o, -\mathbf{k}o\rangle$ and we measure a photon in the $-\mathbf{k}$ path using a single-photon detector that clicks when a photon hits it. Because these two photons are identical in their properties, it is the case that I will know exactly when the undetected photon will arrive at some point downstream on its path. This assumes the photons have some localization in time, but that is a reasonable assumption once we take finite pump bandwidth and phase-matching physics into account. This is considered a photon "on demand" and is useful when one wants photons at controlled times with controlled properties for experiments.

### 5.2.2 Generation of squeezed states by degenerate parametric down-conversion

We see that we have in principle a two-mode Bogoliubov transformation for each pair $(\mathbf{k}_{1,\perp}\omega_1, \mathbf{k}_{2,\perp}\omega_2)$. This corresponds to a type of two-mode squeezing which leads to a reduced variance in joint quadratures involving creation and annihilation operators of both modes. Typically of more interest (because it is simpler to use) is single-mode squeezing. In Unit 4, we wrote squeezing as a single-mode Bogoliubov transformation of the form $a \to \mu a + \nu a^\dagger$. Let us use the above treatment to anticipate when this happens. We wish to arrange things such that $\mathbf{k}_\perp = 0$ allows for phase matching with $\omega_1 = \omega_2 = \omega_3/2$. This can be done using for example type-I phase-matching $(e \to o, o)$ in a negative uniaxial crystal. If we assume the phase-matching bandwidth is small, then the $\nu$ coefficient will only be significant for the mode concentrated around $\mathbf{k}_\perp = 0, \omega = \omega_3/2$. Defining $a = a_{0,\omega_3/2}$, we then have from Eq. II.9 (adapted to discrete operators) with $\Delta k_z = 0$

$$a \to \mu a + \nu a^\dagger, \text{with } \mu = \cosh \lambda L, \nu = ie^{i\phi} \sinh \lambda L, \tag{5.2.23}$$

where we have written $A_3 = |A_3|e^{i\phi}$. We note that in this simple case, this could have also been obtained quite directly from the degenerate parametric amplifier. The classical degenerate parametric amplifier with collinear waves has classical equations of motion

$$\frac{dA_1}{dz} = \frac{2i\omega_1 d_{\text{eff}}}{n_1 c} A_3 A_1^* e^{-i\Delta k z};\ \frac{dA_3}{dz} = \frac{i\omega_3 d_{\text{eff}}}{n_2 c} A_1^2 e^{i\Delta k z}. \tag{5.2.24}$$

In the non-depleted pump approximation, we can treat $A_3$ as constant, which yields in the phase-matched case

$$A_1(L) = A_1(0)\cosh\lambda L + ie^{i\phi}A_1^*(0)\sinh\lambda L, \tag{5.2.25}$$

with $\lambda = \frac{2\omega_1 d_{\text{eff}}|A_3(0)|}{n_1 c}$ and $A_3 = |A_3|e^{i\phi}$ which agrees with the above reduction of the multimode case. This is to be expected. However, it illustrates nicely the ideas of operator correspondence. From this description, we further see that the quadrature variances are given by

$$(\Delta X_\theta)^2 = \frac{1}{2}\left|\cosh(\lambda L)e^{-i\theta} - ie^{i\phi}\sinh(\lambda L)e^{i\theta}\right|^2, \tag{5.2.26}$$

which has a minimum for $\theta = -\frac{\phi}{2} - \pi/4$ and a maximum for $\theta = -\frac{\phi}{2} + \pi/4$. The minimum variance is $\frac{1}{2}e^{-2\lambda L}$, which tells us that the squeezing goes exponentially in the length of propagation and the nonlinearity. In practice, because nonlinearities tend to be weak, it can be hard to get $\lambda L$ significant without significant intensities, which lead to other complications.

## 5.3 Squeezing in the optical parametric oscillator

An approach to getting squeezing with lower powers is to put the parametric crystal in a cavity surrounded by two highly reflective mirrors. This is nothing other than the parametric oscillator we discussed in Unit 2. This allows the parametrically generated light to make multiple passes inside the cavity before it escapes. The way to think about this is with a quantity called finesse, which roughly is "the number of bounces" before light significantly leaks out. It is on the order of $1/T$ where $T$ is the combined transmission coefficient of the two mirrors. For example, if we only have 1% transmission per pass, you expect on the order of 100 bounces, which is as if the second-order nonlinear crystal were 100 times longer. That's obviously helpful as it allows us to get away with a smaller pump power (which makes $\lambda$ smaller). This makes the parametric amplifier a workhorse for squeezed light generation.

Let us work out the theory of squeezed light generation in the parametric oscillator. In some sense, the analysis is quite similar. The basic unit of dynamics in the oscillator is a round trip. In a round trip, light (1) passes through the parametric crystal, undergoing a Bogoliubov transformation, (2) bounces off of the first mirror, (3) propagates backwards to the second mirror (no parametric gain at this stage), and (4) bounces off of the second mirror. In what follows, we will assume that the parametric gain is weak on a single-round trip level which implies that $\nu$ associated with a single pass satisfies $|\nu|^2 \ll 1$. We will also assume that the mirrors have high reflectivity and only transmit a small amount of the field in the cavity, which we need in order to have high finesse. Importantly, transmission is loss from the perspective of the intracavity light and so we need to make sure to account for the quantum noise that is coupled in from outside of the cavity.

The assumption that all of these effects are weak will allow us to approximate each round trip by a differential operator transformation. This allows us to write a differential equation in time for the intracavity field. Beamsplitter-type relations will relate the intracavity field operators to the output field operators that we observe directly. Let us proceed.

1. Propagation through the nonlinear crystal of length $L$ leads to the transformation $a \to \mu a + \nu a^\dagger$ with $\mu = \cosh(\lambda L) \approx 1 + \frac{1}{2}(\lambda L)^2$ and $\nu = ie^{i\phi}\sinh(\lambda L) \approx ie^{i\phi}\lambda L$.

2. Reflection off of the first mirror, with complex scattering coefficients $r_1, t_1$. The resulting map is $a \to -r_1 a + i t_1 b_{\text{in},1}$ where $b_{\text{in},1}$ is the field just outside of the mirror. The "in" notation denotes that the $b$ is an "input" field: it couples into the cavity. We may approximate $|r_1| = \sqrt{1-|t_1|^2} \approx 1 - \frac{1}{2}|t_1|^2$ since by assumption $|t_1|^2 \ll 1$.

3. The backward pass we will assume does nothing. At this point I should mention that the field should pick up a propagation phase of the form $2k_z d$ after a round-trip. To make life simple, I will assume that the resonance condition is met for the frequency we are considering, and so $2k_z d = 2m\pi$ with $m$ an integer. We will also assume that the reflection phases have already been taken into account in the resonance condition and therefore treat all $r, t$ as real.

4. The second reflection simply maps $a \to -r_2 a + i t_2 b_{\text{in},2}$.

The combined effect of these transformations for the $s$th round trip can be written as

$$a_s \to a_{s+1} = r_1 r_2 \mu a_s + r_1 r_2 \nu a_s^\dagger - i t_1 r_2 b_{\text{in},1,s} + i t_2 b_{\text{in},2,s}. \tag{5.3.1}$$

To get to a differential equation, we would like to compute

$$\frac{a_{s+1} - a_s}{\tau} = \frac{1}{\tau}\left( (r_1 r_2 \mu - 1) a_s + r_1 r_2 \nu a_s^\dagger - i t_1 r_2 b_{\text{in},1,s} + i t_2 b_{\text{in},2,s} \right), \tag{5.3.2}$$

where $\tau$ is the round-trip time. Now, we will approximate this as

$$\frac{1}{\tau}\left( (r_1 r_2 - 1) a_s + i(\lambda L) a_s^\dagger - i t_1 b_{\text{in},1,s} + i t_2 b_{\text{in},2,s} \right), \tag{5.3.3}$$

where we have used $|r_{1,2}| \approx 1$ and neglected quadratic in $\lambda L$ terms since $\lambda L \ll 1$. Let us take $r_{1,2}$ and $t_{1,2}$ real. Using $|r_{1,2}| =\approx 1 - \frac{1}{2}|t_{1,2}|^2$, we get

$$\frac{1}{\tau}\left( -\frac{1}{2}(|t_1|^2 + |t_2|^2) a_s + i(\lambda L) a_s^\dagger - i t_1 b_{\text{in},1,s} + i t_2 b_{\text{in},2,s} \right). \tag{5.3.4}$$

It is standard to define the cavity decay rate $\gamma_i = \frac{|t_i|^2}{\tau}$ and $\gamma = \gamma_1 + \gamma_2$. We also define $g = i\lambda L/\tau$. This leaves us with

$$\frac{a_{s+1} - a_s}{\tau} \approx -\frac{1}{2}\gamma a_s + g a_s^\dagger - i\sqrt{\gamma_1}\frac{b_{\text{in},1,s}}{\sqrt{\tau}} + i\sqrt{\gamma_2}\frac{b_{\text{in},2,s}}{\sqrt{\tau}}. \tag{5.3.5}$$

To proceed, we should try and understand these input fields a bit better. The basic idea is that each time the cavity field sees the mirror, fresh vacuum is brought in. Clearly you expect vacuum to have no correlations and so that vacuum modes associated with different round trips are independent of each other. This can be expressed via the commutator

$$\left[ \frac{b_{\text{in},i,s}}{\sqrt{\tau}}, \frac{b_{\text{in},j,s'}^\dagger}{\sqrt{\tau}} \right] = \delta_{ij}\frac{\delta_{s,s'}}{\tau}. \tag{5.3.6}$$

Let us now convert this discrete difference equation into a differential equation. Formally, this occurs through the limit $\tau \to 0$ and is expected to be fine provided the operators $a, a^\dagger$ don't

change too much over one round trip. This creates the replacements $a_s \to a(t), b_{\text{in},i,s} \to b_{\text{in},i}(t)$. Let us perform a change in notation

$$\frac{b_{\text{in},i}(t)}{\sqrt{\tau}} \to b_{\text{in},i}(t). \tag{5.3.7}$$

We may now express the commutator in the $\tau \to 0$ limit as

$$[b_{\text{in},i}(t), b^\dagger_{\text{in},j}(t)] = \delta_{ij}\delta(t-t'). \tag{5.3.8}$$

From now on, if we write $b_{\text{in}}$ it will imply delta-function commutator normalization unless otherwise stated. Further defining $F_1 = -i\sqrt{\gamma_1}b_{\text{in},1}$, $F_2 = i\sqrt{\gamma_2}b_{\text{in},2}$ and $F = F_1 + F_2$, we have

$$\dot{a} = -\frac{1}{2}\gamma a + g a^\dagger + F. \tag{5.3.9}$$

which is the form that we will work with for the remainder of our analysis of the optical parametric oscillator. For completeness, we write also the equation of motion for $a^\dagger$, given by

$$\dot{a}^\dagger = -\frac{1}{2}\gamma a^\dagger + g^* a + F^\dagger. \tag{5.3.10}$$

This equation says that the time-evolution of the intracavity field is given by the combined influence of loss, parametric gain, and in-coupling of vacuum from the outside of the cavity. We see that this in-coupling is related to the decay which is no surprise given our treatment of the beamsplitter. The notation $F$ is used for this term to make apparent that this term plays the role of a Langevin force analogous to that of Brownian motion in statistical mechanics. There is a zero mean driving term which adds a required noise to preserve commutation relations[8]. We will refer to these terms as Langevin forces in what follows.

Before solving this equation, we should discuss the fields that *emanate* from the cavity. Clearly, the only fields we can see are the ones outside of the cavity[9]. It follows immediately from the beamsplitter relations we developed that the output fields satisfy

$$b_{\text{out},i}(t) = -r b_{\text{in},i}(t) + \frac{i t_i}{\sqrt{\tau}} a(t) \approx -b_{\text{in},i}(t) + i\sqrt{\gamma_i}a(t). \tag{5.3.11}$$

This equation of motion for the intracavity field operator can be solved by standard (Fourier) techniques. Fourier transforming, we have

$$\begin{aligned} -i\omega a(\omega) &= -\frac{1}{2}\gamma a(\omega) + g a(-\omega)^\dagger + F(\omega) \\ -i\omega a(-\omega)^\dagger &= -\frac{1}{2}\gamma a(-\omega)^\dagger + g^* a(\omega) + F(-\omega)^\dagger, \end{aligned} \tag{5.3.12}$$

where I have used $\int dt\ e^{i\omega t} A(\omega) = (A(-\omega))^\dagger$. This matrix equation is solved by

$$\begin{pmatrix} a(\omega) \\ a(-\omega)^\dagger \end{pmatrix} = \left( \left(\frac{1}{2}\gamma - i\omega\right)^2 - |g|^2 \right)^{-1} \begin{pmatrix} \frac{1}{2}\gamma - i\omega & g \\ g^* & \frac{1}{2}\gamma - i\omega \end{pmatrix} \begin{pmatrix} F(\omega) \\ F(-\omega)^\dagger. \end{pmatrix} \tag{5.3.13}$$

[8] In statistical mechanics, the analogue is that the Langevin force is needed to preserve for example the equilibrium distribution and subsequent results such as the equipartition theorem. This connection between fluctuation and dissipation is the so-called fluctuation-dissipation theorem. The addition of a Langevin term to our cavity equation can be seen as a quantum-mechanical manifestation of the fluctuation-dissipation theorem.

[9] It is possible to probe intracavity fields, but this is not too important for what we are doing.

### 5.3.1 Squeezing of light outside of the cavity

As discussed previously, what we actually measure are the output fields, so let us compute those. We will consider a slightly simpler situation in which the cavity is perfectly reflective on one end, in which case there is only one output, taken without loss of generality to be the "1" port. We will also, without loss of generality, take the pump phase such that $g$ is real. A different phase of $\phi$ just changes which quadrature gets squeezed, which you are encouraged to check. We will go into Fourier domain, introducing frequency-domain input and output operators with commutator

$$[b_{\text{in}}(\omega), b_{\text{in}}^\dagger(\omega')] = 2\pi\delta(\omega - \omega'). \tag{5.3.14}$$

The output field operator in frequency space is given by

$$\chi^{bb}(\omega) = i\sqrt{\gamma_1}a(\omega) - b_{\text{in}}(\omega) \equiv \chi^{bb}(\omega)b_{\text{in}}(\omega) + \chi^{bb^\dagger}(\omega)b_{\text{in}}(-\omega)^\dagger, \tag{5.3.15}$$

where

$$\chi^{bb}(\omega) = i\sqrt{\gamma_1}\left(\frac{-i\sqrt{\gamma_1}\left(\frac{1}{2}\gamma_1 - i\omega\right)}{\left(\frac{1}{2}\gamma_1 - i\omega\right)^2 - g^2}\right) - 1 = \frac{\frac{\gamma_1^2}{4} + \omega^2 + g^2}{\left(\frac{1}{2}\gamma_1 - i\omega\right)^2 - g^2}. \tag{5.3.16}$$

and

$$\chi^{bb^\dagger}(\omega) = -\frac{\gamma_1 g}{\left(\frac{1}{2}\gamma_1 - i\omega\right)^2 - g^2}. \tag{5.3.17}$$

The relevant observables to compute are understood as follows. You can think of the output of the cavity as a one-dimensional transmission line that supports the propagation of photons at any frequency. There is a time-dependent quadrature, which can be written as $X_\theta(t) = \frac{b(t)e^{i\theta} + b^\dagger(t)e^{-i\theta}}{\sqrt{2}}$. If we were to probe this light by homodyne detection, we would see a time-dependent current $i(t)$ which is proportional to $X_\theta(t)$. Typically we probe squeezing by looking at the power spectral density of the current which is a measure of the variance of an RF component of the current. This variance is proportional to a quantity we will call $S_\theta^{XX}$, the squeezing spectrum, which is a type of power spectral density over a quantity $X(\omega) = b(\omega)e^{-i\theta} + b(-\omega)^\dagger e^{i\theta}$ via [10]:

$$S_\theta^{XX} = \langle X(\omega)^\dagger X(\omega)\rangle. \tag{5.3.18}$$

To evaluate this, let's write

$$X(\omega) = (\chi^{bb}(\omega)b_{\text{in}}(\omega) + \chi^{bb^\dagger}(\omega)b_{\text{in}}(-\omega)^\dagger)e^{-i\theta} + (\chi^{bb*}(-\omega)b_{\text{in}}(-\omega)^\dagger + \chi^{bb^\dagger *}(-\omega)b_{\text{in}}(\omega))e^{i\theta} \tag{5.3.19}$$

and regroup terms as

$$X(\omega) = (\chi^{bb}(\omega)e^{-i\theta} + \chi^{bb^\dagger *}(-\omega)e^{i\theta})b_{\text{in}}(\omega) + (\chi^{bb^\dagger}(\omega)e^{-i\theta} + \chi^{bb*}(-\omega)e^{i\theta})b_{\text{in}}(-\omega)^\dagger. \tag{5.3.20}$$

Assuming vacuum statistics for the input fields, it follows that the squeezing spectrum is given by

$$S_\theta^{XX}(\omega) = \left|\chi^{bb^\dagger}(\omega)e^{-i\theta} + \chi^{bb*}(-\omega)e^{i\theta}\right|^2. \tag{5.3.21}$$

[10] We have dropped the overall factor of 1/2 here as squeezing is in any way a claim about a variance relative to vacuum.

For our particular system, we have

$$S_\theta^{XX}(\omega) = \Big| \frac{-\gamma_1 g e^{-i\theta} + (\frac{\gamma_1^2}{4} + \omega^2 + g^2) e^{i\theta}}{\left(\frac{1}{2}\gamma_1 - i\omega\right)^2 - g^2} \Big|^2 \tag{5.3.22}$$

The minimum and maximum of the squeezing spectra occur when $\theta = 0, \pi/2$. The minimum is for $\theta = 0$ and the corresponding spectrum is

$$S_0^{XX} = \frac{\left(\frac{\gamma_1}{2} - g\right)^2 + \omega^2}{\left(\frac{\gamma_1}{2} + g\right)^2 + \omega^2}, \tag{5.3.23}$$

while in the orthogonal quadrature, the spectrum is given as:

$$S_{\pi/2}^{XX} = \frac{\left(\frac{\gamma_1}{2} + g\right)^2 + \omega^2}{\left(\frac{\gamma_1}{2} - g\right)^2 + \omega^2}. \tag{5.3.24}$$

These are reciprocals of each other: $S_0^{XX} S_{\pi/2}^{XX} = 1$. Interestingly, we see that as $g \to \frac{\gamma_1}{2}$, the squeezing spectrum goes to zero for $\theta = 0$, corresponding to infinite squeezing at $\omega = 0$, while the squeezing spectrum diverges for the orthogonal quadrature. Note that $\omega$ is a frequency *relative* to the carrier (our Heisenberg equations do not have the $-i\omega_0 a$ term associated with the cavity resonance frequency). Therefore, the interpretation is that light coming out of the cavity at the resonance frequency can have extremely large squeezing, and indeed the largest squeezing levels created in the lab are using optical parametric oscillators.

The condition of $g = \frac{\gamma_1}{2}$ corresponds to the threshold for parametric oscillation. Recall in Unit 2 we showed that when the gain in the OPO exceeds the cavity losses, we expect a oscillating steady-state in the cavity. While if the gain is below the losses, the steady state should be at zero amplitude (modulo quantum corrections from parametric fluorescence). The bifurcation between decay and oscillation behaviors is at the threshold when the gain equals the loss. The threshold should be re-examined for this case since in Unit 2 we had not analyzed a degenerate oscillator directly[11]. Recall that the resonance condition is that the fields reproduce themselves after a round-trip. Ignoring the Langevin terms, that condition, following the logic of Eq. (III.5) requires that the matrix

$$-\frac{1}{2}\gamma_1 I + \begin{pmatrix} 0 & g \\ g^* & 0 \end{pmatrix} \tag{5.3.25}$$

is non-invertible, which requires a vanishing of the determinant:

$$\frac{\gamma_1^2}{4} - |g|^2 \implies |g| = \frac{\gamma_1}{2}. \tag{5.3.26}$$

Another way to see that this is the threshold is to look at the eigenvalues of the homogeneous part of the linear system of equations (Eqs. (III.9) and (III.10)) which shows that perturbations grow without bound if $g > \gamma_1/2$.

[11] That said, the condition can be found from our Unit 2 analysis by taking the relevant threshold to be for the doubly-resonant case ($\lambda L = \sqrt{\ell_i \ell_s}$) and taking both resonances to be the same so that $\ell = 1 - r \approx \frac{1}{2}|t|^2 = \frac{1}{2}\gamma_1 \tau$. Taking $\lambda L = g\tau$ then reproduces the same condition.

### 5.3.2 Intracavity quadrature statistics

Although in practice we only have access to what's outside of the cavity, it is instructive to compute the intracavity light properties, and so we will start there. Our aim is to compute quadrature variances:

$$(\Delta X)^2 = \frac{1}{2}\langle (a + a^\dagger)^2\rangle, (\Delta P)^2 = -\frac{1}{2}\langle (a - a^\dagger)^2\rangle, \tag{5.3.27}$$

where I have used that the means vanish. The means vanish because they are proportional to the input field, which is assumed to be in the vacuum state. Had we taken a coherent state input, the mean would not vanish, as you expect (if I shine light into the cavity from the outside, there will in general be a nonzero intracavity field).

Let's start with $X$. This can be written in terms of the Fourier fields via

$$X(t) = \int \frac{d\omega}{2\pi} e^{-i\omega t} X(\omega), \tag{5.3.28}$$

where $X(\omega) = \frac{1}{\sqrt{2}}\left(a(\omega) + a(-\omega)^\dagger\right)$. Therefore

$$(\Delta X)^2 = \int \frac{d\omega}{2\pi}\frac{d\omega'}{2\pi} e^{-i(\omega+\omega')t}\langle X(\omega)X(\omega')\rangle. \tag{5.3.29}$$

Let us now consider the quantity $\langle X(\omega)X(\omega')\rangle$ which is given by:

$$\left\langle \left(\chi^{XF}(\omega)F(\omega) + \chi^{XF^\dagger}(\omega)F(-\omega)^\dagger\right)\left(\chi^{XF}(\omega')F(\omega') + \chi^{XF^\dagger}(\omega')F(-\omega')^\dagger\right)\right\rangle \tag{5.3.30}$$

where I have defined constants $\chi^{XF}, \chi^{XF^\dagger}$ multiplying the Langevin forces. Assuming the input field to be vacuum, we have that the $FF$, $F^\dagger F$, and $F^\dagger F^\dagger$ expectation values vanish (as $F$ is proportional to an oscillator annihilation operator $b_{\text{in}}$). The remaining term is $\langle F(\omega)F(-\omega)^\dagger\rangle = \langle [F(\omega), F(-\omega)^\dagger]\rangle = \gamma_1 \times 2\pi\delta(\omega + \omega')$, which follows from $[b_{\text{in}}(\omega), b_{\text{in}}^\dagger(\omega')] = 2\pi\delta(\omega - \omega')$. We see then that

$$(\Delta X)^2 = \gamma_1 \int \frac{d\omega}{2\pi}\chi^{XF}(\omega)\chi^{XF^\dagger}(-\omega). \tag{5.3.31}$$

Using

$$\chi^{XF}(\omega) = \frac{1}{\sqrt{2}}\frac{\frac{1}{2}\gamma_1 - i\omega + g}{\left(\frac{1}{2}\gamma_1 - i\omega\right)^2 - g^2} = \frac{1}{\sqrt{2}}\frac{1}{\frac{1}{2}\gamma_1 - i\omega - g} \tag{5.3.32}$$

and

$$\chi^{XF^\dagger}(-\omega) = \frac{1}{\sqrt{2}}\frac{\frac{1}{2}\gamma_1 + i\omega + g}{\left(\frac{1}{2}\gamma_1 + i\omega\right)^2 - g^2} = \frac{1}{\sqrt{2}}\frac{1}{\frac{1}{2}\gamma_1 + i\omega - g}, \tag{5.3.33}$$

we have

$$(\Delta X)^2 = \frac{1}{2}\int \frac{d\omega}{2\pi}\frac{1}{\left(\frac{1}{2}\gamma_1 - g\right)^2 + \omega^2} = \frac{1}{4}\frac{\gamma_1}{\frac{1}{2}\gamma_1 - g} = \frac{1}{2}\frac{1}{1 - \frac{2g}{\gamma_1}}. \tag{5.3.34}$$

The $P$ variance follows immediately from its definition as

$$(\Delta P)^2 = \frac{1}{2}\frac{1}{1 + \frac{2g}{\gamma_1}}. \tag{5.3.35}$$

In this case, at threshold, we have that $(\Delta X)^2 \to \infty$ and $(\Delta P)^2 \to \frac{1}{4}$, corresponding only to 3 dB of squeezing. This is very different from what happens outside of the cavity, where the squeezing (in the $X$-like quadrature) diverges. The zeroing of the noise for the field at the outside essentially comes from an interference of vacuum fluctuations reflecting off of the outside of the cavity and light from the cavity escaping. The light from the cavity however experiences no such interference.

## 5.4 Quantum noise dynamics in third-order nonlinear systems

At this stage, we have spent a considerable amount of time analyzing second-order nonlinearity. It turns out that many of these phenomena can also be replicated by third-order nonlinear systems. Mirroring the treatment of third-order systems we pursued in Unit 3, we will focus on effects resulting from the intensity-dependent refractive index, taking fields whose interesting spectral content is localized to some center frequency $\omega_1$. This allows us to expand the dispersion relation around this frequency, getting an ultrashort pulse propagation equation like Eq. I.15. Unlike the case we analyzed for the bulk crystal, we will content ourselves with unidirectional propagation along a waveguide (we won't worry about beam propagation here), in which case, we may simply take $\mathbf{E}^{(+)}(\mathbf{r},\omega) = \mathbf{E}^{(+)}(z,\omega)$. We will also assume all interesting effects occur along only one polarization, in which case we may consider a scalar field $E^{(+)}(z,\omega)$. It is useful to express this field in terms of an envelope via $E^{(+)}(z,\omega) = A^{(+)}(z,\omega)e^{ik(\omega_1)z}$. In time-domain, we could write $E^{(+)}(z,t) = A^{(+)}(z,t)e^{ik(\omega_1)z - i\omega_1 t}$ and the total field operator is written as

$$E(z,t) = A^{(+)}(z,t)e^{ik(\omega_1)z - i\omega_1 t} + A^{(-)}(z,t)e^{-ik(\omega_1)z + i\omega_1 t}. \tag{5.4.1}$$

A very similar derivation to the classical case then yields the Heisenberg equation

$$\partial_z \tilde{A}_1^{(+)}(z,t) = \sum_{m=1}^{\infty} \frac{i^{m+1}\beta_m}{m!}\partial_t^m \tilde{A}_1^{(+)}(z,t) + \frac{i\omega_1^2}{2\epsilon_0 c^2 k(\omega_1)}\tilde{P}_1^{(+)}(z,t). \tag{5.4.2}$$

The relevant polarization at third-order is given by

$$\tilde{P}_1^{(+)}(z,t) = \epsilon_0 \chi^{(3)} E(z,t)^3. \tag{5.4.3}$$

Since we are only interested in the part of the polarization near frequency $\omega$, we may take only the terms in $E^3$ that multiply $e^{ik(\omega_1)z - i\omega_1 t}$. Those terms are

$$\left(\tilde{A}_1^{(+)}(z,t)\tilde{A}_1^{(+)}(z,t)\tilde{A}_1^{(-)}(z,t) + \tilde{A}_1^{(+)}(z,t)\tilde{A}_1^{(-)}(z,t)\tilde{A}_1^{(+)}(z,t) + \tilde{A}_1^{(-)}(z,t)\tilde{A}_1^{(+)}(z,t)\tilde{A}_1^{(+)}(z,t)\right). \tag{5.4.4}$$

To simplify this, it is useful to have commutation relations for $A^{(+)}(z,t)$ and $A^{(-)}(z',t')$. We will take this also as an opportunity to simplify and move to some more standard notation. Clearly:

$$[A^{(+)}(z,t), A^{(-)}(z',t')] = [E^{(+)}(z,t), E^{(-)}(z',t')]e^{-ik(\omega_1)(z-z') + i\omega_1(t-t')}. \tag{5.4.5}$$

The electric field operator, in our quasi-1D, scalar case can be written as an expansion in plane waves as

$$E^{(+)}(z,t) = i\int_{\Delta k} \frac{dk}{2\pi}\mathcal{E}(k)a(k,t)e^{ikz - i\omega_k t}, \tag{5.4.6}$$

where $\mathcal{E}(k)$ is a constant and $a(k)$ is defined such that $[a(k), a^\dagger(k')] = 2\pi\delta(k-k')$. The constant $\mathcal{E}(k)$, neglecting group velocity dispersion, can be written as

$$\mathcal{E}(k) = \sqrt{\frac{\hbar\omega(k)}{2\epsilon_0 n^2 A}}, \tag{5.4.7}$$

with $n$ an effective mode index and $A$ an effective mode area. We restrict the mode expansion to a wavenumber bandwidth $\Delta k$ since this simplified description of the field may not be valid everywhere. Therefore, the commutator can be written as

$$C^E(z-z', t-t') = [E^{(+)}(z,t), E^{(-)}(z',t')] = \int_{\Delta k} \frac{dk}{2\pi} |\mathcal{E}(k)|^2 e^{ik(z-z') - i\omega_k(t-t')}. \tag{5.4.8}$$

The equal time commutator is given by

$$C^E(z-z', 0) = \int_{\Delta k} \frac{dk}{2\pi} |\mathcal{E}(k)|^2 e^{ik(z-z')}. \tag{5.4.9}$$

Let us suppose that the bandwidth $\Delta k$ is such that the variations of $|\mathcal{E}(k)|^2$ can be neglected, being set to $\mathcal{E}(k_0)|^2$. Then we get (assuming the limits are from $-\Delta k/2$ to $\Delta k/2$:

$$C^E(z-z', 0) \approx |\mathcal{E}(k_0)|^2 \frac{\Delta k}{2\pi} \text{sinc}\left(\frac{\Delta k}{2}(z-z')\right). \tag{5.4.10}$$

In other words, we can think of our spatial field as being composed of "localized photon modes" of width $1/\Delta k$. if for example $\Delta k = k_0/10$ then for $\lambda = 1\mu$m, the localization scale is roughly $10\lambda \sim 10\mu$m. For $z = z'$ one simply gets $C^E(0,0) = |\mathcal{E}(k_0)|^2/L_{\text{ph}}$ where $L_{\text{ph}} = 2\pi/\Delta k$. We may use this to simplify the sum of cubic envelope operators, writing it as

$$3\tilde{A}_1^{(-)}(z,t)\tilde{A}_1^{(+)}(z,t)\tilde{A}_1^{(+)}(z,t) + 3A^{(+)}(z,t)C^E(0,0). \tag{5.4.11}$$

The second term is an effective renormalization of the wavevector. The more rigorous way to deal with it is to absorb it as a wavevector shift in the Heisenberg equation. The less rigorous way is to ignore it. However, for the circumstances we will consider, this term will be very small compared to the cubic term since we will typically work with fields that have many photons in them (we need strong fields for bulk nonlinearities to be significant in effect). The commutator term is proportional to the nonlinearity induced by a single photon and so is neglected in what follows.

Using this simplification our Heisenberg equation may now be written as

$$\partial_z \tilde{A}_1^{(+)}(z,t) = \sum_{m=1}^{\infty} \frac{i^{m+1}\beta_m}{m!} \partial_t^m \tilde{A}_1^{(+)}(z,t) + \frac{3i\omega_1\chi^{(3)}}{2cn_1(\omega)} \tilde{A}_1^{(-)}(z,t)\tilde{A}_1^{(+)}(z,t)^2. \tag{5.4.12}$$

One may see that this is in direct correspondence with the classical pulse propagation equations that we wrote in Unit 3. The envelope operator has dimensions of electric field. In what follows, let us define a dimensionless operator which has commutation relations we expect for continuum field operators. Define

$$A^{(+)}(z,t) = \sqrt{\frac{\hbar\omega_1}{2\epsilon_0 n^2 A}}\psi(z,t); A^{(-)}(z,t) = \sqrt{\frac{\hbar\omega_1}{2\epsilon_0 n^2 A}}\psi^\dagger(z,t) \tag{5.4.13}$$

Then, in the limit of infinite bandwidth $C^{\psi}(0,0) = [\psi(z,t), \psi^{\dagger}(z,t)] = \delta(z-z')$, and our operator-valued pulse propagation equation becomes

$$\partial_z \psi(z,t) = \sum_{m=1}^{\infty} \frac{i^{m+1}\beta_m}{m!} \partial_t^m \psi(z,t) + i\kappa \psi^{\dagger}(z,t)\psi^2(z,t), \tag{5.4.14}$$

with $\kappa = \frac{3\hbar\omega_1^2\chi^{(3)}}{4\epsilon_0 c n_1^3 A}$, where $n_1 = n(\omega_1)$. In the presence of dispersion, where the group velocity different from the phase velocity, $\kappa = \frac{3\hbar\omega_1^2\chi^{(3)}v_g}{4\epsilon_0 n_1^2 c^2 A}$. Finally, as is standard in pulse propagation, we express these equations in a co-moving frame introducing the usual variables $z' = z, t' = t - \beta_1 z$ which leaves us with

$$\partial_{z'} \psi(z',t') = \sum_{m=2}^{\infty} \frac{i^{m+1}\beta_m}{m!} \partial_t^m \psi(z',t') + i\kappa \psi^{\dagger}(z',t')\psi^2(z',t'). \tag{5.4.15}$$

The commutator at equal $z$ can *approximately* be written as

$$C^{\psi}(0, t_1' - t_2') \approx \frac{\Delta k}{2\pi} \mathrm{sinc}\left(\frac{v_g \Delta k}{2}(t_1' - t_2')\right). \tag{5.4.16}$$

For a sufficiently large bandwidth, we may write this as $\frac{1}{v_g}\delta(t-t')$. The interpretation of $\psi^{\dagger}\psi$ is a photon number density (photon number per unit length). It is more convenient to rescale our $\psi$ to be a flux of photons (photons per unit time). This is done be rescaling $\psi$ by $\psi \to \sqrt{v_g}\psi$ and $\kappa/v_g \to \kappa$ with $v_g$ the group velocity. This leaves us with our final equation:

$$\partial_{z'} \psi(z',t') = \sum_{m=2}^{\infty} \frac{i^{m+1}\beta_m}{m!} \partial_t^m \psi(z',t') + i\kappa \psi^{\dagger}(z',t')\psi^2(z',t'). \tag{5.4.17}$$

In what follows, we will consider the idealized limit in which $C^{\psi}(0, t-t') = \delta(t-t')$. Before proceeding, it useful to note that the $\kappa$ parameter is related to the $\gamma$ that we know from the classical pulse-propagation equation from Unit 3 with power-normalized fields. The relation is $\kappa = \gamma\hbar\omega$. This makes sense as $\kappa$ is the constant for photon-number-per-unit-time-normalized fields while $\gamma$ is the constant for energy-per-unit-time normalized fields.

### 5.4.1 Squeezing by self-phase modulation

Let us analyze the very simplest nonlinear effect which is self-phase modulation. In other words, we will set our dispersive terms to zero in the quantum pulse-propagation equation, which leaves us with

$$\partial_z \psi(z,t') = i\kappa \psi^{\dagger}(z,t')\psi^2(z,t'), \tag{5.4.18}$$

where I have taken $z' \to z$ since their meaning is the same. This equation is exactly solvable. It uses the fact that

$$\partial_z \psi^{\dagger}(z,t')\psi(z,t') = \psi^{\dagger}(z,t')(i\kappa\psi^{\dagger}(z,t')\psi^2(z,t')) - (i\kappa\psi^{\dagger 2}(z,t')\psi(z,t'))\psi(z,t') = 0. \tag{5.4.19}$$

Therefore, we may write our Heisenberg equation as

$$\partial_z \psi(z,t') = i\kappa(\psi^{\dagger}(0,t')\psi(0,t'))\psi(z,t'), \tag{5.4.20}$$

which looks like something that can be exponentiated. That said however, we must be very careful when exponentiating operator equations like this. If $\psi^\dagger(0,t')\psi(0,t')$ were a $c$-number, we could exponentiate directly. Let us however make a trial solution of the form

$$\psi(z,t') = e^{i\kappa z\psi^\dagger(0,t')\psi(0,t')}\psi(0,t'). \tag{5.4.21}$$

Since $[e^{i\kappa z\psi^\dagger(0,t')\psi(0,t')}, \psi^\dagger(0,t')\psi(0,t')] = 0$, it immediately follows that upon differentiation by $z$ the $i\kappa\psi^\dagger(0,t')\psi(0,t')$ that is pulled down can be pulled to the left of the exponent, satisfying our differential equation.

Let us analyze the physics of this transformation, which can be seen as a photon-number dependent phase rotation (very much like how we think of classical self-phase modulation). To expose the physics a bit more cleanly, let us work with unit-commutator creation and annihilation operators. We see that each time-slice of the pulse behaves independently, so let us define a time-slice operator

$$a \equiv \sqrt{\Delta t}\psi(t), \tag{5.4.22}$$

where $\Delta t$ is some temporal bin width. This allows us to associate the self-phase modulation with a transformation

$$a \to e^{i\theta a^\dagger a}a, \tag{5.4.23}$$

where $\theta = \kappa z \times \frac{1}{\Delta t} = \gamma z \times \frac{\hbar\omega}{\Delta t}$. The factor $\frac{\hbar\omega}{\Delta t}$ can be seen as the power carried by a single photon pulse of temporal width $\Delta t$. We are mostly introducing this time-bin as a formal device to map our pulse problem into a single-mode problem. The transformation above $a \to e^{i\theta a^\dagger a}a$ is one that appears in more general contexts such as third-order nonlinear media inside optical cavities with a single well-defined resonance mode (associated with operator $a$). In such circumstances, we typically associate an interaction picture Hamiltonian $H = \frac{\hbar K}{2}a^{\dagger 2}a^2$ with corresponding Heisenberg equation (for the interaction picture field operator) $\dot{a} = -iKa^\dagger a^2$ which has the same formal solution as above. Here, if you want to take this time-slice operator seriously, you should treat $\Delta t$ as the regularization time associated with the field commutator $C^\psi(0,0)$.

This transformation can be enacted exactly. But we will first consider a simpler limit in which the input is in a coherent state $|\alpha\rangle$ where $|\alpha|^2 \gg 1$. Then, we can treat the operator $a = \alpha + \delta a$ where $\delta a$ represents the quantum fluctuations of the operator and $\alpha$ is the mean field. This is exactly equivalent to saying that we take expectations with respect to the vacuum state but treat our initial operator as displaced: $a \to a + \alpha$. But since we used equality rather than $\to$, we should call the operator on the right-hand side something different, and so we call it $\delta a$. In any event, it has the interpretation as being the quantum fluctuations on top of an otherwise classical field of a coherent state. In this case, we expect the argument of the exponent to be small. This is because typically in nonlinear optics we deal with nonlinear phases on the order of $2\pi$. But if we consider

$$\theta a^\dagger a = \theta\left(|\alpha|^2 + (\alpha^*\delta a + \alpha\delta a^\dagger) + \delta a^\dagger\delta a\right), \tag{5.4.24}$$

we see that the classical nonlinear phase corresponds to the first term. The terms linear and quadratic in fluctuations are much weaker. If $\alpha = \sqrt{n}$ where $n$ is a characteristic number of photons inside the field then we see the linear terms are smaller by $\sqrt{n}$ and the quadratic terms by $n$. In typical contexts involving typical materials, the number of photons needed

for a $2\pi$ nonlinear phase shift is much greater than one. At the very leading order, we could neglect both the linear and quadratic terms, but then we would learn no information about quantum statistics. The leading quantum behavior is maintained by keeping the terms linear in $\delta a$, leaving us with

$$a \to e^{i\theta a^\dagger a} a \approx e^{i\theta|\alpha|^2}\left(1 + i\theta(\alpha^*\delta a + \alpha\delta a^\dagger)\right)(\alpha + \delta a). \tag{5.4.25}$$

In this, I used the fact that if $\theta|\alpha|^2$ is of order 1, then the linear terms are much smaller than one. By a similar logic, we can drop all terms in the remaining expression which are higher-order-than-linear, which gives us

$$a \to e^{i\theta|\alpha|^2}\left(\alpha + \left(1 + i\theta|\alpha|^2\right)\delta a + i\theta\alpha^2\delta a^\dagger\right). \tag{5.4.26}$$

This transformation, which is equivalent to a Bogoliubov transformation, acting on an initial coherent state, gives a displaced squeezed state. Let us try and understand this. First, since the mean-field (the average field) is much larger than the fluctuation operators, we can equate averages and fluctuations separately. The transformation of the average field is

$$\alpha \to e^{i\theta|\alpha|^2}\alpha, \tag{5.4.27}$$

which is the usual SPM transformation. The fluctuations transform as

$$\delta a \to \mu\delta a + \nu\delta a^\dagger, \text{where } \mu = \left(1 + i\theta|\alpha|^2\right)e^{i\theta|\alpha|^2}, \nu = i\theta\alpha^2 e^{i\theta|\alpha|^2} \tag{5.4.28}$$

We immediately see that $|\mu|^2 - |\nu|^2 = 1$ as we expect for the Bogoliubov transformation. The $\zeta$ parameter for the transformation, or equivalently $|r|$ and $\phi$ depend on the initial field amplitude, and so the squeezed quadrature changes as we scale up the amplitude! To explicitly connect the transformation above, written in terms of operators, in terms of squeezing followed by displacement, let us consider for some operator $b$ the transformation

$$b' = D^\dagger(\alpha)S^\dagger(\zeta)bS(\zeta)D(\alpha) = D^\dagger(\alpha)(\mu b + \nu b^\dagger)D(\alpha) = \mu(b + \alpha) + \nu(b^\dagger + \alpha^*), \tag{5.4.29}$$

where $\mu, \nu$ are determined from $\zeta$ in the usual way. Now define fluctuation operators via $\delta b' = b' - \langle b'\rangle$. Then, we have that the output fluctuation operators are given by

$$\delta b' = \mu(b + \alpha) + \nu(b^\dagger + \alpha^*) - (\mu\alpha + \nu\alpha^*) = \mu b + \nu b^\dagger \tag{5.4.30}$$

where we have assumed that the initial state is the vacuum state without loss of generality. For the initial vacuum state, since the mean is zero, $b$ and $\delta b$ coincide. Therefore we have

$$\delta b' = \mu\delta b + \nu\delta b^\dagger, \tag{5.4.31}$$

describing the transformation we found from self-phase modulation. It is important to understand that since this state is nothing other than a displaced squeezed state, quadrature variances work the same way as for the squeezed vacuum state. The state is just shifted in phase-space such that it has a mean complex amplitude.

The squeezing operation can also be understood in terms of a phase space picture. If we consider our coherent state as a point cloud of random complex amplitudes with some variance, then some of these initial amplitudes correspond to larger photon number, and accumulate phase more rapidly due to SPM. This leads to a deformation of the symmetric distribution into an ellipse for small deformations, yielding squeezing. For larger phase shifts where the linearization approximation breaks down, one gets a more "meniscus shaped" distribution and we should instead appeal to an exact quantum calculation.

### 5.4.2 Track 2: Generation of Schrodinger cat states by Kerr nonlinearity

When linearization breaks down, we can get more "exotic" quantum states than the squeezed state. For example, we can get a superposition of two coherent states of the same magnitude displacement. When that displacement is large, it corresponds to a superposition of two macroscopic light beams called a Schrodinger cat state in analogy to the eponymous cat which is in a superposition of alive and dead. To see how this occurs, we want to work in the Schrodinger picture. As mentioned previously, the interaction picture evolution of a state due to self-modulation is generated by a Hamiltonian $H_K = \frac{\hbar K}{2} a^{\dagger 2} a^2 = \frac{\hbar K}{2}(N^2 - N)$ where $N = a^\dagger a$.

The time-evolution is then (all states are now in interaction picture)

$$|\psi'\rangle = e^{-i\theta N} e^{i\theta N^2} |\psi(0)\rangle \tag{5.4.32}$$

where $\theta = -Kt/2$. Suppose that we have a coherent state as our initial state. Then, employing its photon number representation, we see that

$$|\psi'\rangle = e^{-\frac{1}{2}|\alpha|^2} \sum_{n=0}^{\infty} e^{i\theta n^2} \frac{(\alpha e^{-i\theta})^n}{\sqrt{n!}} |n\rangle. \tag{5.4.33}$$

Let us consider what happens for $\theta = \pi/2$. You can convince yourself in this case that $e^{in^2\theta} = e^{i\pi/2}$ for every odd $n$, and 1 for every even $n$. We may thus write

$$e^{in^2\theta} = \frac{1}{\sqrt{2}} e^{i\frac{\pi}{4}} + (-1)^n e^{-i\frac{\pi}{4}}. \tag{5.4.34}$$

Therefore it immediately follows that the resulting state is

$$|\psi'\rangle = \frac{1}{\sqrt{2}} \left( e^{i\frac{\pi}{4}} |\alpha e^{-i\theta}\rangle + e^{-i\frac{\pi}{4}} | - \alpha e^{-i\theta}\rangle \right), \tag{5.4.35}$$

corresponding to a superposition of two coherent states. It is worth noting that this state has fairly exotic properties but is incredibly hard to realize using bulk nonlinearities because it requires the phase shift of a single photon being on the order of 1, which for typical fibers would require millions of meters (which ignores loss and many other aspects of reality that would compromise cat-state generation. However, these very same ideas in the context of superconducting qubits, which present strong single-photon Kerr-like nonlinearity at *microwave* frequencies, are applicable, and cat states have been generated for use in quantum computing schemes.

## 5.5 Track 2: Semiclassical picture of quantum noise

*Note: This material is largely copied from Nature Photonics 19.7 (2025): 751-757.*
It is hard not to notice that there is a strong correspondence between the operator valued Heisenberg equations and the classical nonlinear equations that we have studied thus far. In fact, when there are strong pump fields involved that can be treated classically, we often end up with linear operator equations which are formally identical to the classical equations with

commutator constraints. In this so-called linearization regime, it turns out that there is a general and simple way to get variances of arbitrary observables in terms of derivatives of classical nonlinear equations of motion. This connection is called quantum sensitivity analysis and we derive it here.

Consider a system of bosons described by a set of annihilation and creation operators $a_i, a_i^\dagger$ where $i$ is a generalized index, labeling not only different modes for a given boson, but also different types of bosons. We'll denote the vector of operators as $\mathbf{a}, \mathbf{a}^\dagger$ for compactness. For example, in a case where light interacts with phonons, as well as atoms capable of absorbing the light, the boson operators may label photon modes, phonon modes, and the effective bosonic modes which describe an absorbing material.

The equation of motion for the operators can be written schematically as:

$$\begin{aligned}\dot{\mathbf{a}} &= F(\mathbf{a}, \mathbf{a}^\dagger) \\ \dot{\mathbf{a}}^\dagger &= F^\dagger(\mathbf{a}, \mathbf{a}^\dagger),\end{aligned} \tag{5.5.1}$$

where $F$ is a generic operator function that produces the right-hand side of the Heisenberg equations of motion corresponding to the system Hamiltonian.

In the case where the number of bosons in the initial state is large, the quantum dynamics can be well approximated by the lowest-order fluctuations on top of the mean-field dynamics [**?**]. This "linearization" approximation proceeds by expressing the operators as

$$\mathbf{a} = \boldsymbol{\alpha} + \delta\mathbf{a}, \tag{5.5.2}$$

where $\boldsymbol{\alpha} \equiv \langle \mathbf{a} \rangle$, plugging it into the Heisenberg equation, and neglecting terms of higher order than linear in $\delta\mathbf{a}$. The equation of motion for the mean fields can be written as

$$\dot{\boldsymbol{\alpha}} = F(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*), \tag{5.5.3}$$

where the $c$-number function $F(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*)$ corresponds to replacing all operators in $F(\mathbf{a}, \mathbf{a}^\dagger)$ by c-numbers. These equations will be nothing other than the classical equations of motion for the system. Meanwhile, the equation of motion for the fluctuations can be expressed as

$$\delta\dot{a}_i = \sum_j \frac{\partial F_i}{\partial \alpha_j}\delta a_j + \sum_j \frac{\partial F_i}{\partial \alpha_j^*}\delta a_j^\dagger \tag{5.5.4}$$

where, importantly, the derivatives are with respect to the classical ($c$-number) function. The equation of motion for the creation operators just follows from conjugation.

The solution to these equations can be expressed as a Bogoliubov transformation, as:

$$\delta a_i(t) = \sum_j \mu_{ij}\delta a_j(0) + \nu_{ij}\delta a_j^\dagger(0). \tag{5.5.5}$$

In what follows, we will make a new connection, showing that the $\mu, \nu$ are in fact derivatives of the classical equations of motion with respect to the initial conditions. To see this, let us consider when we take the classical equation of motion Eq. 5.5.3 and evaluate the change in the solution when the initial conditions are varied. Assuming that a variation in the initial

conditions, $\delta\boldsymbol{\alpha}$ leads to a sufficiently small change in the output fields, we may linearize the equations, writing them as

$$.\delta\dot{\alpha}_i = \sum_j \frac{\partial F_i}{\partial \alpha_j}\delta\alpha_j + \sum_j \frac{\partial F_i}{\partial \alpha_j^*}\delta\alpha_j^* \tag{5.5.6}$$

This equation is in correspondence with Eq. 5.5.4, and its solution is the same, as Eq. 5.5.4 is a linear equation, and thus is solved identically to a classical equation. In particular, we may write:

$$\delta\alpha_i(t) = \sum_j \mu_{ij}\delta\alpha_j(0) + \nu_{ij}\delta\alpha_j^\dagger(0), \tag{5.5.7}$$

where $\mu, \nu$ are identical to the quantum-mechanical case. But, by construction, we may also write

$$\delta\alpha_i(t) = \sum_j \frac{\partial\alpha_i(t)}{\partial\alpha_j(0)}\delta\alpha_j(0) + \frac{\partial\alpha_i(t)}{\partial\alpha_j^*(0)}\delta\alpha_j^*(0), \tag{5.5.8}$$

allowing us to identify

$$\mu_{ij} = \frac{\partial\alpha_i(t)}{\partial\alpha_j(0)}, \nu_{ij} = \frac{\partial\alpha_i(t)}{\partial\alpha_j^*(0)}, \tag{5.5.9}$$

completing the proof.

We now use this result to connect the calculation of quantum noise in multimode systems of light and matter to adjoint methods for solving numerical differential equations. Consider an observable $\delta X$ which is linear in fluctuation operators $\delta a_i$ and $\delta a_i^\dagger$:

$$\delta X = \sum_i c_i\delta a_i + d_i\delta a_i^\dagger. \tag{5.5.10}$$

For a wide range of observables operators $X$, the corresponding fluctuation operators $\delta X = X - \langle X\rangle$ will take the form above. In particular, for any operator function of creation and annihilation operators, we may write $\delta X = \nabla_\alpha X(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*)\cdot\delta\mathbf{a} + \nabla_{\alpha^*}X(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*)\cdot\delta\mathbf{a}^\dagger + O(\delta a\delta a, \delta a\delta a^\dagger, ...)$. Therefore, we can identify $c_i = \partial X/\partial\alpha_i$ and $d_i = \partial X/\partial\alpha_i^*$. For states with Gaussian correlations, odd-order correlators vanish, a variance such as $\langle(\delta X)^2\rangle$ will have terms of second-order in fluctuation operators and terms of fourth-order in fluctuation operators: assuming we keep terms up to quadratic order in the fluctuation operators. For a Gaussian state, the fourth-order correlators will factorize in terms which go as the square of second-order correlators. As an example of an operator, consider $X = n = a^\dagger a$. Then, so long as $|\alpha|^2 \gg \langle\delta a\delta a\rangle, \langle\delta a\delta a^\dagger\rangle, ...$, the linearized approximation will be valid. An equivalent order-of-magnitude comparison is obtained by comparing $n^2$ with the variance $(\Delta n)^2$, as would be expected from the fact that linearization is valid when fluctuations are small.

In terms of the results of the previous section, we may write

$$\begin{aligned}
\delta X(t) &= \sum_{ij}\left[c_i\frac{\partial\alpha_i(t)}{\partial\alpha_j(0)} + d_i\frac{\partial\alpha_i^*(t)}{\partial\alpha_j(0)}\right]\delta a_j(0) + \left[c_i\frac{\partial\alpha_i(t)}{\partial\alpha_j^*(0)} + d_i\frac{\partial\alpha_i^*(t)}{\partial\alpha_j^*(0)}\right]\delta a_j^\dagger(0) \\
&= \sum_j \frac{\partial X(t)}{\partial\alpha_j(0)}\delta a_j(0) + \frac{\partial X(t)}{\partial\alpha_j^*(0)}\delta a_j^\dagger(0) \\
&\equiv \left[\delta\boldsymbol{a}\cdot\frac{\partial}{\partial\boldsymbol{\alpha}} + \delta\boldsymbol{a}^\dagger\cdot\frac{\partial}{\partial\boldsymbol{\alpha}^*}\right]X(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*)
\end{aligned} \tag{5.5.11}$$

We have used the chain rule to express the result in terms of the derivative of the classical observable with respect to initial conditions. In the last line, and in what follows throughout most of this Supplement, we will omit explicit indication of time-zero quantities. We also briefly note that in the main text, for intuitive ease, we have replaced the time-zero label by "in" and the time-$t$ label by "out". Further, in the last line, we have introduced the notation $X(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*)$, which refers to the value of the observable $X$, calculated *classically*, assuming some initial conditions $\boldsymbol{\alpha}, \boldsymbol{\alpha}^*$. While classically, the initial conditions and their conjugates would not be independent, here, we take them as independent, since they refer to fluctuations in $a$ and $a^\dagger$, which quantum-mechanically have a nonvanishing commutator.

Since this expression applies to any observable, we may now write a compact relation for the variance in an arbitrary observable, in an arbitrary coupled system of light and matter degrees of freedom, in this linearization approximation. This relation is what we refer to as the quantum optical law of total variance. Within this approximation, the variance of $X$, $(\Delta X)^2 = \langle (\delta X)^2 \rangle$, is expressed as a quadratic form:

$$\begin{aligned} (\Delta X)^2 &= v^T C v, \text{where} \\ v &= \left( \frac{\partial X}{\partial \boldsymbol{\alpha}} \ \frac{\partial X}{\partial \boldsymbol{\alpha}^*} \right)^T \\ C &= \begin{pmatrix} \langle \delta \mathbf{a} \delta \mathbf{a} \rangle & \langle \delta \mathbf{a} \delta \mathbf{a}^\dagger \rangle \\ \langle \delta \mathbf{a}^\dagger \delta \mathbf{a} \rangle & \langle \delta \mathbf{a}^\dagger \delta \mathbf{a}^\dagger \rangle \end{pmatrix} \end{aligned} \tag{5.5.12}$$

The correlation matrix $C$ which multiplies the gradients $v$ is constructed from the statistics of the initial field, which allows for straightforward inclusion of the effects of excess noise, multimode correlations (e.g., entanglement), and phase-sensitive correlations (e.g., from squeezed states of light). This reformulation shows that the noise is in fact largely understood in terms of the gradient of the classical transformation of the light with respect to the initial conditions. This enables one to predict and understand the dynamics of fluctuations and noise based on the classical understanding of nonlinear optical effects developed over recent years. That said, one key aspect of this framework that goes "beyond" many classical studies is that: in this framework, one must also study the sensitivity of the system to dark modes, as it is these which often limits the noise performance in the presence of interactions.

### 5.5.1 Examples of quantum sensitivity analysis

In this section, we apply the quantum sensitivity analysis formulae to two cases we have already studied: single-mode parametric amplification, and self-phase modulation of pulses.

#### Single-mode parametric amplifier

In the derivation of quantum sensitivity analysis, we assumed that the operator $X$ could be non-trivially linearized. That requires that the gradients $\nabla_\alpha X$ do not vanish. In what follows, we present a simple case where the gradient does vanish, and show that our framework can still capture the correct result. The solution will be to use the more general expression relating the Bogoliubov coefficients to the classical Jacobian. As an example, we consider the intensity fluctuations of squeezed vacuum. To describe fluctuations of squeezed vacuum using

our framework, the first step is to identify and solve the corresponding classical model with a general initial condition. The equations of motion are

$$\begin{aligned}\dot{\alpha} &= \Omega\alpha^* \\ \dot{\alpha}^* &= \Omega^*\alpha.\end{aligned} \tag{5.5.13}$$

The solution is

$$\begin{pmatrix}\alpha(t)\\ \alpha^*(t)\end{pmatrix} = \exp\left[\begin{pmatrix}0 & \Omega t\\ \Omega^* t & 0\end{pmatrix}\right]\begin{pmatrix}\alpha(0)\\ \alpha^*(0)\end{pmatrix} = \begin{pmatrix}\cosh\left(|\Omega|t\right)\alpha(0) + \sinh(|\Omega|t)e^{i\phi}\alpha^*(0)\\ \sinh(|\Omega|t)e^{-i\phi}\alpha(0) + \cosh\left(|\Omega|t\right)\alpha^*(0)\end{pmatrix}, \tag{5.5.14}$$

where we have defined $\Omega = |\Omega|e^{i\phi}$. According to Eq. (S5), we can write the fluctuation operators as

$$\delta a(t) = \mu\delta a(0) + \nu\delta a^\dagger(0), \tag{5.5.15}$$

where $\mu = \partial\alpha(t)/\partial\alpha(0) = \cosh r$ and $\nu = \partial\alpha(t)/\partial\alpha^*(0) = e^{i\phi}\sinh r$, where $r = |\Omega|t$. This is of course the correct Bogoliubov transformation expected quantum mechanically noting that since $\alpha(t) = 0$, $a = \delta a$. Let us now consider a quadrature operator $X_\varphi = ae^{i\varphi} + a^\dagger e^{-i\varphi}$. With this normalization, the quadrature variance in the vacuum state is one. Let us now ask about the squeezing of an initially injected vacuum state. From the quantum optical law of total variance, the quadrature variance is simply:

$$(\Delta X_\varphi)^2 = \left|\frac{\partial X_\varphi(t)}{\partial a(0)}\right|^2 = \left|\cosh\left(|\Omega|t\right)e^{i\varphi} + \sinh(|\Omega|t)e^{-i\phi}e^{-i\varphi}\right|^2, \tag{5.5.16}$$

which is the known result.

Now, let us consider the intensity or photon number fluctuations. A direct application of Eq. (S12) yields zero for the case of squeezed vacuum generation, where $\alpha(0), \alpha^*(0) = 0$, since the derivative of $n(t) = \alpha^*(t)\alpha(t)$ with respect to initial conditions will lead to a sum of terms, each of which are linear in $\alpha(t)$ or $\alpha^*(t)$. This is a simple consequence of the fact that the gradient $\nabla_\alpha n$ is vanishing, meaning that $n$ cannot be approximated as linear in fluctuation operators for this initial condition. To get the mean photon number and the photon number fluctuations, one simply computes $\langle n\rangle = \langle a^\dagger a\rangle$ and $\langle n^2\rangle$ the standard way, which will reproduce the known result (see e.g. [**?**]).

We now use QSA to describe *displaced* squeezed vacuum. Classically, one can model this by taking the parametric amplifier relation above and mixing that light through a beamsplitter with another classical light beam called a local oscillator [12]. The resulting transformation is

$$\alpha = \beta + \mu\alpha(0) + \nu\alpha^*(0). \tag{5.5.17}$$

The gradient of the photon number at $\alpha(0), \alpha^*(0) = 0$ is given by $n = \alpha^*\alpha$, $\partial n/\partial\alpha(0) = \beta^*\mu + \beta\nu^*$, and so the intensity fluctuations in response to a vacuum state input are $(\Delta n)^2 = |\beta^*\mu + \beta\nu^*|^2 = |\beta|^2\left(e^{2r}\cos^2\theta + e^{-2r}\sin^2\theta\right)$, for the case where $\mu = \cosh r, \nu = \sinh r$ and defining $\beta = |\beta|e^{i\theta}$. This is the known result when $|\beta|^2 \gg |\nu|^2$ (see [**?**]), which is the limit assumed by linearization.

[12] For the displaced squeezing to be valid, and for us to neglect vacuum fluctuations of the intense field, it needs to be the case that the transmission of the amplifier output is approximately one (so the intense field is mostly attenuated, but still very intense compared to the number of photons in the amplifier output).

**Self-phase modulation of pulses**

For a pulse in a medium experiencing pure self-phase modulation without temporal dispersion, the classical nonlinear equation of motion is

$$\partial_z \alpha(z,t) = i\gamma \alpha^*(z,t)\alpha^2(z,t), \tag{5.5.18}$$

where $\alpha(z,t)$ is the envelope of the electric field of the pulse as a function of distance along the fiber $z$ and time-delay relative to the center of the pulse (defined as time zero). Dimensions of $\alpha$ are chosen such that $|\alpha|^2 dt$ represents the number of photons carried in a time-span $dt$ of the pulse. As is standard in ultrafast nonlinear optics, this equation is in the frame of reference which co-propagates with the pulse at its group velocity $v_g$ [**?**]. The quantity $\gamma$ is related to the nonlinear propagation phase. In particular, the pulse envelope, after a propagation distance $L$ in the fiber, is given by:

$$\alpha(L,t) = e^{i\theta\alpha^*(0,t)\alpha(0,t)}\alpha(0,t), \tag{5.5.19}$$

where $\theta \equiv \gamma L$, which follows from the fact that $|\alpha|^2(z,t) = |\alpha|^2(0,t)$. It follows immediately from these equations, as well as Eq. (S5) and (S9), that

$$\delta a(L,t) = \int ds \ \left[\frac{\partial\alpha(L,t)}{\partial\alpha(0,s)}\delta a(0,s) + \frac{\partial\alpha(L,t)}{\partial\alpha^*(0,s)}\delta a^\dagger(0,s)\right], \tag{5.5.20}$$

where

$$\frac{\partial\alpha(L,t)}{\partial\alpha(0,s)} = \delta(t-s)\left(1 + i\theta|\alpha(0,t)|^2\right)e^{i\theta|\alpha|^2(0,t)} \equiv \mu(s)\delta(t-s), \tag{5.5.21}$$

and

$$\frac{\partial\alpha(L,t)}{\partial\alpha^*(0,s)} = \delta(t-s)\left(i\theta\alpha(0,t)^2\right)e^{i\theta|\alpha|^2(0,t)} \equiv \nu(s)\delta(t-s), \tag{5.5.22}$$

where we have defined $\mu$ and $\nu$ for ease of notation. This relation is in exact agreement with the standard treatment based on an exact solution of the Heisenberg equations.

# Appendices

# Appendix A

# Appendix A: Maxwell's equations in materials

*Maxwell's equations in polarizable media. Scalar and vector potentials. Maxwell equations as eigenvalue problems. Green's functions (general). Green's functions for the Maxwell equations. Green's functions in terms of mode expansions. Electromagnetic energy in nondispersive and dispersive electromagnetic media.*

## A.1 Maxwell's equations in materials

The goal of any problem in classical electrodynamics is to find the time-evolution of the electromagnetic fields under a prescribed distribution of charges and currents. The time-evolution is governed by the Maxwell equations. In frequency-domain, the equations take a much simpler form, which we will almost exclusively use. As you saw in the previous section, the Maxwell equations in frequency domain are given by

$$\nabla \cdot \epsilon(\mathbf{r},\omega)\mathbf{E}(\mathbf{r},\omega) = \frac{\rho(\mathbf{r},\omega)}{\epsilon_0} \tag{A.1.1}$$

$$\nabla \cdot \mathbf{B}(\mathbf{r},\omega) = 0 \tag{A.1.2}$$

$$\nabla \times \mathbf{E}(\mathbf{r},\omega) = i\omega\mathbf{B}(\mathbf{r},\omega) \tag{A.1.3}$$

$$\nabla \times \mathbf{H}(\mathbf{r},\omega) = \mathbf{J}(\mathbf{r},\omega) - i\omega\epsilon_0\epsilon(\mathbf{r},\omega)\mathbf{E}(\mathbf{r},\omega), \tag{A.1.4}$$

where $\epsilon(\mathbf{r},\omega)$ is the frequency-dependent permittivity of the previous chapter. $\rho$ and $\mathbf{J}$ are *free* charge and current densities. Bound charges and currents, as we discussed in the previous section, are captured by the permittivity. As a warning, we will often leave out position and frequency functional dependences. They are implicit.

### A.1.1 Maxwell's equations for the potentials

When we formulate quantum electrodynamics for electrons interacting with quantized fields in materials, we will go through the vector and scalar potential, as it makes evident the number of dynamical degrees of freedom needed to describe the electromagnetic field. Therefore,

we shall formulate Maxwell's equation for the potentials, as well as introduce the so called *generalized Coulomb gauge*, which is key to the quantization of electromagnetic fields in media. Recall that in electromagnetism, we can define a vector potential $\mathbf{A}$ and a scalar potential $\phi$ such that $\mathbf{E} = -\nabla\phi - \partial_t\mathbf{A}$ and $\mathbf{B} = \nabla \times \mathbf{A}$. In frequency domain, $\partial_t\mathbf{A} \to -i\omega\mathbf{A}$. The Maxwell equations for the potentials in frequency domain then read:

$$\nabla \cdot (-i\omega\epsilon(\mathbf{r},\omega)\mathbf{A}(\mathbf{r},\omega) + \epsilon(\mathbf{r},\omega)\nabla\phi(\mathbf{r},\omega)) = -\frac{\rho(\mathbf{r},\omega)}{\epsilon_0} \tag{A.1.5}$$

$$\nabla \times \nabla \times \mathbf{A}(\mathbf{r},\omega) = \mu_0\mathbf{J}(\mathbf{r},\omega) - \frac{i\omega}{c^2}\epsilon(\mathbf{r},\omega)\left(i\omega\mathbf{A}(\mathbf{r},\omega) - \nabla\phi(\mathbf{r},\omega)\right). \tag{A.1.6}$$

You will see that the second and third Maxwell equations are trivially satisfied and require no additional effort on our part. We may omit them. Next, we shall make use of *gauge invariance* of Maxwell's equations. In particular, if we perform the following transformations on the potentials: $\phi \to \phi - i\omega\chi$ and $\mathbf{A} \to \mathbf{A} - \nabla\chi$, the electric and magnetic fields remain unaffected. Thus, the electric and magnetic fields are the "physical" fields. Thus, we can choose any gauge that makes convenient the solution of the remaining equations. We will make use of the *generalized Coulomb gauge* $\nabla \cdot \epsilon\mathbf{A} = 0$. You are encouraged to prove that one may always find a gauge transformation to satisfy this condition, provided that the gauge function satisfies $\nabla \cdot \epsilon\nabla\chi = 0$. In this gauge, the remaining Maxwell equations, after some re-arrangement, satisfy:

$$\nabla \cdot \epsilon(\mathbf{r},\omega)\nabla\phi(\mathbf{r},\omega) = -\frac{\rho(\mathbf{r},\omega)}{\epsilon_0} \tag{A.1.7}$$

$$\left(\nabla \times \nabla \times -\epsilon(\mathbf{r},\omega)\frac{\omega^2}{c^2}\right)\mathbf{A}(\mathbf{r},\omega) = \mu_0\mathbf{J}(\mathbf{r},\omega) + \frac{i\omega}{c^2}\epsilon(\mathbf{r},\omega)\nabla\phi(\mathbf{r},\omega). \tag{A.1.8}$$

I will comment that an advantage of these equations is that it is clear that there are only three degrees of freedom needed to describe the electromagnetic fields: one scalar potential, and *two* components of the vector potential. The third component of the vector potential is determined from the other two by the generalized Coulomb gauge condition. But the information in these equations is equivalent to the information in the Maxwell equations for the fields. For example we can easily get the electric field from these previous two equations. Plugging in $\mathbf{A} = \frac{\mathbf{E}+\nabla\phi}{i\omega}$, we see that

$$\left(\nabla \times \nabla \times -\epsilon(\mathbf{r},\omega)\frac{\omega^2}{c^2}\right)\mathbf{E}(\mathbf{r},\omega) = i\omega\mu_0\mathbf{J}(\mathbf{r},\omega). \tag{A.1.9}$$

The magnetic field is simply $\frac{\nabla\times\mathbf{E}}{i\omega}$. Let us now understand the general properties of the solutions to these equations.

### A.1.2 General properties of Maxwell's equations in the absence of sources

Consider Maxwell's equations in the absence of sources: $\mathbf{J} = 0$ and $\rho = 0$. The scalar potential equation is solved by $\phi = c$ with $c$ a constant. As we can always perform a gauge transformation that removes this constant, we may simply say $\phi = 0$. Thus, the only equation of interest is that for the vector potential, which simplifies to

$$\nabla \times \nabla \times \mathbf{A}(\mathbf{r},\omega) = \epsilon(\mathbf{r},\omega)\frac{\omega^2}{c^2}\mathbf{A}(\mathbf{r},\omega) \tag{A.1.10}$$

The equation for the vector potential on the other hand does have non-zero solutions for $\omega \neq 0$. For example, if $\epsilon = 1$, then a plane transverse wave of wavevector magnitude $k$ satisfies this equation when $\omega = ck$. In what follows, we will consider a *non-dispersive*, but *inhomogeneous* dielectric medium, so that $\epsilon(\mathbf{r}, \omega) = \epsilon(\mathbf{r})$. We will also only consider *lossless* media for the present, such that $\epsilon$ is real-valued. A number of powerful general results emerge simply in this case. The resulting equation is a *generalized eigenproblem*, in which an operator $(\nabla \times \nabla \times)$, acting on a (vector) function $(\mathbf{A})$, gives that same function, up to an eigenvalue $(\frac{\omega^2}{c^2})$, and a function $(\epsilon)$. In the absence of this additional function, this would be a standard eigenproblem. Nevertheless, many results from standard eigenproblems carry over with minimal modification. Here, we enumerate the basic properties of this generalized eigenproblem, and its consequences. These consequences will be critical in the quantization of the electromagnetic field.

To derive basic properties of the Maxwell eigenproblem, we will need to define an inner product between vector functions. In particular, we define

$$\langle \mathbf{X}, \mathbf{Y} \rangle \equiv \int d^3r \; \mathbf{X}^* \cdot \mathbf{Y}. \tag{A.1.11}$$

You are encouraged to confirm that this inner product satisfies the axioms of inner-product spaces. Given this inner product, we may now simply prove (the first two of) the following properties of the Maxwell equations, its eigenfunctions (or eigenmodes) $\mathbf{F}_n(\mathbf{r}) \equiv \mathbf{A}(\mathbf{r}, \omega_n)$, and its corresponding eigenfrequencies $\omega_n$:

1. **The eigenproblem is Hermitian.** In particular $\langle \mathbf{X}, \nabla \times \nabla \times \mathbf{Y} \rangle = \langle \nabla \times \nabla \times \mathbf{X}, \mathbf{Y} \rangle$. To prove this: let us start on the right side of the equality and integrate-by-parts our way over to the left hand side. Recall that $\int d^3r \; \mathbf{A} \cdot \nabla \times \mathbf{B} = -\oint dS \; \hat{n} \cdot (\mathbf{A} \times \mathbf{B}) + \int d^3r \; (\nabla \times \mathbf{A}) \cdot \mathbf{B}$. In everything we will be doing, we will take boundary conditions at infinity such that either: the fields vanish, or they are periodic at the boundary. In either case, this makes the boundary term zero. Thus, we may focus only on the second term. The integration by parts then tells us that the curl moves over (with no change in sign, due to the antisymmetric properties of cross products). Thus, the second curl also moves over for free, and it is then clear that $\langle \mathbf{X}, \nabla \times \nabla \times \mathbf{Y} \rangle = \langle \nabla \times \nabla \times \mathbf{X}, \mathbf{Y} \rangle$, which means $\nabla \times \nabla \times$ is a Hermitian operator.

2. **The eigenvalues are real if $\epsilon$ is positive.** Consider $\langle \mathbf{F}_n, \nabla \times \nabla \times \mathbf{F}_n \rangle = \frac{\omega_n^2}{c^2} \langle \mathbf{F}_n, \epsilon \mathbf{F}_n \rangle$. By integration by parts, we may write the left-hand side as $\langle \nabla \times \mathbf{F}_n, \nabla \times \mathbf{F}_n \rangle = \int d^3r \; |\nabla \times \mathbf{F}_n|^2 > 0$. If $\epsilon \geq 0$, then $\langle \mathbf{F}_n, \epsilon \mathbf{F}_n \rangle > 0$, $\omega_n^2 > 0$, and thus $\omega_n$ is real. Both positive and negative roots are valid solutions, and correspond to positive and negative frequencies in the frequency-domain Fourier expansion of the electromagnetic field we considered at the beginning of the lecture.

3. **The eigenfunctions can be made orthonormal according to a modified inner product**. Eigenfunctions of different eigenvalues are manifestly orthogonal (and then normalized to be orthonormal) with respect to a new inner product $(\mathbf{X}, \mathbf{Y}) \equiv \int d^3r \; \epsilon \mathbf{X}^* \cdot \mathbf{Y}$. To see this, simply consider the following inner product:

$$\langle \mathbf{F}_m, \nabla \times \nabla \times \mathbf{F}_n \rangle = \frac{\omega_n^2}{c^2} \langle \mathbf{F}_m, \epsilon \mathbf{F}_n \rangle = \frac{\omega_n^2}{c^2} \langle \epsilon \mathbf{F}_m, \mathbf{F}_n \rangle. \tag{A.1.12}$$

By Hermiticity under the standard inner product, it is the case that this must also equal

$$\langle \nabla \times \nabla \times \mathbf{F}_m, \mathbf{F}_n \rangle = \frac{\omega_m^2}{c^2} \langle \epsilon \mathbf{F}_m, \mathbf{F}_n \rangle. \tag{A.1.13}$$

It is thus immediately seen that if $\omega_m \neq \omega_n$, then $(\mathbf{F}_m, \mathbf{F}_n) = \int d^3r \ \epsilon \mathbf{F}_m^* \cdot \mathbf{F}_n = 0$. Degenerate eigenfunctions can be made orthonormal by the Gram-Schmidt procedure, just as in quantum mechanics.

4. **The eigenfunctions span the space of divergenceless functions.** Proving this requires functional analysis, so we will invoke the result instead. Consider a divergenceless (transverse) function $\mathbf{X}$, such that $\nabla \cdot \mathbf{X} = 0$. Then it may be expanded in terms of the eigenfunctions of the Maxwell's equations, up to a factor of $\epsilon$, via:

$$\mathbf{X} = \sum_n c_n \epsilon \mathbf{F}_n, \tag{A.1.14}$$

with $c_n$ the expansion coefficients, given by $c_m = \int d^3r \ \epsilon \mathbf{F}_m^* \cdot \mathbf{F}_n$. You can see that the divergence of each individual term is zero (from the generalized Coulomb gauge condition), so that the divergence of the overall function is zero.

It then follows that we may write an any time-dependent vector potential in the generalized Coulomb gauge by using a mode expansion, as

$$\mathbf{A}(\mathbf{r}, t) = \frac{1}{2} \sum_n \alpha_n \mathbf{F}_n(\mathbf{r}) e^{-i\omega_n t} + \alpha_n^* \mathbf{F}_n^*(\mathbf{r}) e^{i\omega_n t}, \tag{A.1.15}$$

where $\alpha_n$ are arbitrary coefficients. In writing this form, we have explicitly separated positive and negative frequencies. We have also defined the expansion coefficients with a factor of one-half, so that the right-hand side may also be written as $\text{Re}\left[\sum_n \alpha_n \mathbf{F}_n(\mathbf{r}) e^{-i\omega_n t}\right]$. Keep in mind the expansion of Equation A.1.15, as it will be the central object of study in the canonical quantization of the electromagnetic field. Before moving on, note that for the electric field, one has the exact same eigenproblem in the absence of currents and charges, and the corresponding modes have the same properties (as they are the same modes).

Now that we have a framework for working with the modes of Maxwell's equations in arbitrary media, we can consider how these fields are produced in the first place – through currents and charges. This will reveal the important phenomenon of *radiation*, which we will treat primarily through the method of *Green's functions*.

### A.1.3 Maxwell's equations in the presence of charges and currents

In the presence of charges and currents, we must revert back to Equations A.1.7 and A.1.8. We require additional mathematical formalism to proceed, namely the method of Green's functions, which we now introduce.

### A.1.4 Green's functions

Consider an arbitrary linear differential equation of the form:

$$(\mathcal{D}f) = S(\mathbf{r}), \tag{A.1.16}$$

where $\mathcal{D}$ is a differential operator. $S(\mathbf{r})$ is a source. We have written the equation as a scalar equation, but the general considerations here extend to vector and tensor equations. Examples of a differential operator would be $\nabla^2$, $\nabla^2 + c^2$, with $c$ a constant, $\nabla^4$, $\frac{d}{dx}$, $f(x)\frac{d}{dx}$, etc. The differential operators we'll apply these considerations to are $\nabla \cdot \epsilon \nabla$ and $\nabla \times \nabla \times -\epsilon \frac{\omega^2}{c^2}$. Suppose we now consider the case where the source is a delta function: $S(\mathbf{r}) = \delta(\mathbf{r} - \mathbf{r}')$. We call the corresponding solution of the inhomogeneous equation the Green's function $G(\mathbf{r}, \mathbf{r}')$:

$$(\mathcal{D}G) = \delta(\mathbf{r} - \mathbf{r}'). \tag{A.1.17}$$

If we know the Green's function, then we may solve the differential equation for any source $S(\mathbf{r})$ using linearity. In particular, the solution $f(\mathbf{r})$ can be expressed as an integral of the Green's function with the source as

$$f(\mathbf{r}) = \int d^3r' \; G(\mathbf{r}, \mathbf{r}')S(\mathbf{r}'). \tag{A.1.18}$$

One can easily see why this works by plugging this solution back into Eq. A.1.16 which gives

$$\mathcal{D}f(\mathbf{r}) = \int d^3r' \; \mathcal{D}G(\mathbf{r}, \mathbf{r}')S(\mathbf{r}') = \int d^3r' \; \delta(\mathbf{r} - \mathbf{r}')S(\mathbf{r}') = S(\mathbf{r}), \tag{A.1.19}$$

where we have only used the defining Green's function property of Eq. A.1.17. This means that if we can find the Green's function by solving a differential equation with a delta function source, then we can construct the solution to any source by integrating this source against the Green's function. This is a powerful tool of differential equations in both mathematics and physics.

We now turn to how Green's functions can be used to write solutions to Maxwell's equations. We focus first on the defining equations, and then show how the Green's functions can be expressed in terms of eigenmode expansions. By taking Eq. A.1.7 and replacing source term on the right hand side with a delta function, we can write the equation which defined the Green's function $G_\phi(\mathbf{r}, \omega)$ of the scalar potential as

$$\nabla \cdot \epsilon(\mathbf{r}, \omega) \nabla G_\phi(\mathbf{r}, \omega) = \delta(\mathbf{r} - \mathbf{r}'). \tag{A.1.20}$$

If the solution for $G_\phi(\mathbf{r}, \omega)$ is known, then the scalar potential can be expressed in terms of the charge sources as

$$\phi(\mathbf{r}, \omega) \equiv -\frac{1}{\epsilon_0} \int d^3r' \; G_\phi(\mathbf{r}, \mathbf{r}', \omega)\rho(\mathbf{r}', \omega). \tag{A.1.21}$$

Similarly, replacing the right hand side of Eq. A.1.8 with a delta function, we see that the Green's function $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ should satisfy

$$\left(\nabla \times \nabla \times -\epsilon(\mathbf{r}, \omega)\frac{\omega^2}{c^2}\right) \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}')I. \tag{A.1.22}$$

Unlike the equation for the scalar potential, this is a vector equation with a vector source. As such, the Green's function $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ is necessarily a tensor, and the curls are understood to act on the first index (and first position argument) of the tensor. For this reason, the object $\mathbf{G}$ is often referred to as the *dyadic Green's function*. Consequently, the right hand side is the tensor $\delta(\mathbf{r} - \mathbf{r}')I$, where $I$ is the 3-by-3 identity matrix.

The dyadic Green's function can then be used to write a solution for the vector potential in terms of the current source and scalar potential as

$$\mathbf{A}(\mathbf{r}, \omega) \equiv \int d^3r' \ \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \left( \mu_0 \mathbf{J}(\mathbf{r}', \omega) + \frac{i\omega}{c^2} \epsilon(\mathbf{r}', \omega) \nabla \phi(\mathbf{r}', \omega) \right). \tag{A.1.23}$$

Note here that since $\mathbf{G}$ is a 3-by-3 matrix and the dot here means matrix-vector multiplication. As we will rely on it heavily in the next section, we also want the electric field in terms of Green's functions. Since Eq. A.1.9 expresses the electric field in terms of the same operator, we can easily write

$$\mathbf{E}(\mathbf{r}, \omega) = i\omega\mu_0 \int d^3r' \ \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{J}(\mathbf{r}', \omega). \tag{A.1.24}$$

We are now ready to derive an expression for the Green's function in terms of the eigenmodes $\mathbf{F}_n$ of the source-free problem. This is done by solving Equations A.1.7 and A.1.8 individually, finding the result when the current is a delta function, and combining them to get the electric field. This will enable us to infer the Green's function.

We start with the vector potential. Suppose that the vector potential can be expanded in the basis of modes of the Maxwell eigenproblem, so that

$$\mathbf{A}(\mathbf{r}, \omega) = \sum_n A_n(\omega) \mathbf{F}_n(\mathbf{r}). \tag{A.1.25}$$

By plugging this into Equation (8), we can immediately see that the left-hand side

$$\sum_n \frac{1}{c^2} \left( \omega_n^2 - \omega^2 \right) A_n(\omega) \epsilon(\mathbf{r}, \omega) \mathbf{F}_n(\mathbf{r}) \tag{A.1.26}$$

is divergence-less (transverse), as it should be. This transverse left-hand side is being expanded in a complete set of transverse functions $\epsilon \mathbf{F}_n$, as asserted in the previous section. The right-hand side, being transverse, can be expanded similarly as

$$\sum_n c_n(\omega) \epsilon(\mathbf{r}, \omega) \mathbf{F}_n(\mathbf{r}). \tag{A.1.27}$$

Taking inner products reveals an expression for the coefficients $c_n(\omega)$ as

$$c_n(\omega) = \int d^3r \ \mathbf{F}_n^*(\mathbf{r}) \left( \mu_0 \mathbf{J}(\mathbf{r}, \omega) + \frac{i\omega}{c^2} \epsilon(\mathbf{r}, \omega) \nabla \phi(\mathbf{r}, \omega) \right) = \mu_0 \int d^3r \ \mathbf{F}_n^*(\mathbf{r}) \mathbf{J}(\mathbf{r}, \omega). \tag{A.1.28}$$

The second term is zero. That is because it is the integral of a product of a divergenceless function and a curl-less function. Solving for $A_n$, we find that the vector potential is given by

$$\mathbf{A}(\mathbf{r}, \omega) = \mu_0 c^2 \sum_n \frac{\int d^3r' \ \mathbf{F}_n^*(\mathbf{r}') \cdot \mathbf{J}(\mathbf{r}', \omega)}{\omega_n^2 - \omega^2} \mathbf{F}_n(\mathbf{r}). \tag{A.1.29}$$

Now let's discuss the scalar potential. The scalar potential is simply

$$\phi(\mathbf{r},\omega) = -\frac{1}{\epsilon_0}\int d^3r'\ G_\phi(\mathbf{r},\mathbf{r}',\omega)\rho(\mathbf{r}',\omega). \tag{A.1.30}$$

Combining these together, the electric field is given by

$$\mathbf{E}(\mathbf{r},\omega) = i\omega\mu_0 c^2 \sum_n \frac{\int d^3r'\ \mathbf{F}_n^*(\mathbf{r}')\cdot\mathbf{J}(\mathbf{r}',\omega)}{\omega_n^2-\omega^2}\mathbf{F}_n(\mathbf{r}) + \frac{1}{\epsilon_0}\int d^3r'\ \nabla G_\phi(\mathbf{r},\mathbf{r}',\omega)\rho(\mathbf{r}',\omega) \tag{A.1.31}$$

To construct the Green's function, we need to find the electric field produced by a delta-function current source of frequency $\omega$ and oriented along the $j$ direction, so that $\mathbf{J}(\mathbf{r},\omega) = \hat{e}_j\delta(\mathbf{r}-\mathbf{r}')$. Then we need to divide by $i\omega\mu_0$. For the first term, it is clear what to do. For the second, we need to know what charge density corresponds to a delta function current density, revealing clearly that the charge distribution corresponding to a delta function current is a dipole. From the continuity equation, we see that

$$\rho(\mathbf{r},\omega) = \frac{1}{i\omega}\nabla\cdot\mathbf{J}(\mathbf{r},\omega) \implies \rho_j(\mathbf{r},\omega) = \frac{1}{i\omega}\partial_j\delta(\mathbf{r}-\mathbf{r}'). \tag{A.1.32}$$

It is given by a derivative of a delta function. It follows that

$$\mathbf{G}(\mathbf{r},\mathbf{r}',\omega) = c^2\sum_n \frac{\mathbf{F}_n(\mathbf{r})\mathbf{F}_n^*(\mathbf{r}')}{\omega_n^2-\omega^2+i0^+} + \frac{c^2}{\omega^2-i0^+}\nabla\nabla' G_\phi(\mathbf{r},\mathbf{r}',\omega). \tag{A.1.33}$$

The infinitesimal imaginary parts in the denominators arise from asking: what happens if $\omega = \omega_m$ [1]? It looks like the Green's function diverges. However, in any realistic situation, the divergence is regularized by the fact that any system has at least infinitesimal dissipation. This is called the *principle of limiting absorption*. To apply it, we take $\omega \to \omega - i0^+$, similarly to what we did when studying perturbation theory in quantum mechanics.

[1] or $\omega = 0$, but we're never interested in this situation

# Appendix B

# Appendix B: Lagrangian and Hamiltonian formulation of classical electrodynamics

## B.1 Lagrangian and Hamiltonian formulation of classical electromagnetism

In the previous chapter, we studied the description of the classical electromagnetic field. We reformulated Maxwell's equations in dielectric media in terms of the vector and scalar potential. As argued from the previous section, this reformulation is helpful because it reveals in a transparent way the number of degrees of freedom needed to describe the electromagnetic field. In this chapter, we will *quantize* the combined system of charges and electromagnetic fields. To quantize the system means essentially to turn all observable quantities describing the system into operators. Thus, the positions and velocities of the matter (charges) become operators, as do the electric and magnetic fields. Additionally, we will need to find a Hamiltonian operator which generates the time-evolution of the light-matter system.

We already know how to quantize the charges, as this is just conventional quantum mechanics – thus the essential new element is the quantization of the electromagnetic field. We will see that each mode of the electromagnetic field is associated with a harmonic oscillator.

In general, the strategy to answering these questions is as follows:

1. Start from a Lagrangian whose equations of motion (Euler-Lagrange equations) are classical equations that we already know (the Maxwell equations for the fields, the Newton equations for the charges)

2. Turn the Lagrangian into a Hamiltonian through the standard Legendre transform

3. For matter described by positions and velocities, we quantize as usual, converting the position into the position operator, and the momentum into the momentum operator $\mathbf{p} = -i\hbar\nabla$. For fields, which possess an infinite number of degrees of freedom (to describe the field at all points in space), we will find that degrees of freedom of the EM field can be conveniently cast in terms of the amplitudes of the normal modes of

the field. We will find that the independent field modes act as independent harmonic oscillators, which lead then to a simple and straightforward quantization.

Let us start by finding the Lagrangian of the system of matter and fields. We will do this in three steps: we will first introduce the Lagrangian of a system of matter in an external electromagnetic field. Then, we will introduce the Lagrangian of the electromagnetic field in the absence of charges. Finally, we will derive the Lagrangian describing the interaction between the charges and the fields.

### B.1.1 Lagrangian of matter in an external field

We start by considering the Lagrangian describing a single particle in an external field, described by the scalar potential $\phi(\mathbf{r},t)$ and the vector potential $\mathbf{A}(\mathbf{r},t)$. We are looking to find a Lagrangian $L(\mathbf{r}(t),\dot{\mathbf{r}}(t),t)$ (dots here denote time-derivative) such that when the associated action functional

$$S[\mathbf{r}(t),\dot{\mathbf{r}}(t);t_i,t_f] \equiv \int_{t_i}^{t_f} dt\ L(\mathbf{r}(t),\dot{\mathbf{r}}(t),t) \tag{B.1.1}$$

is stationary, it reproduces the Newtonian equation of motion for the particle in an external electromagnetic field. By stationary, we mean with respect to a change of the functions $\mathbf{r}(t),\dot{\mathbf{r}}(t)$ To minimize the action, let us consider a small variation of the trajectory $\mathbf{r}(t)\to\mathbf{r}(t)+\delta\mathbf{r}(t)$, where $|\delta\mathbf{r}|\ll|\mathbf{r}|$. We can expand the action in this small variation as

$$\begin{aligned} S[\mathbf{r}(t)+\delta\mathbf{r}(t),\dot{\mathbf{r}}(t)+\delta\dot{\mathbf{r}}(t);t_i,t_f] &= \int_{t_i}^{t_f} dt\ L(\mathbf{r}(t)+\delta\mathbf{r}(t),\dot{\mathbf{r}}(t)+\delta\dot{\mathbf{r}}(t),t) \\ &\approx \int_{t_i}^{t_f} dt\ \left(L(\mathbf{r}(t),\dot{\mathbf{r}}(t),t)+\delta\mathbf{r}\cdot\nabla_{\mathbf{r}}L+\delta\dot{\mathbf{r}}\cdot\nabla_{\dot{\mathbf{r}}}L\right). \end{aligned} \tag{B.1.2}$$

The corresponding change in the action is then

$$\delta S = \int_{t_i}^{t_f} dt\ \left(\delta\mathbf{r}\cdot\nabla_{\mathbf{r}}L+\delta\dot{\mathbf{r}}\cdot\nabla_{\dot{\mathbf{r}}}L\right). \tag{B.1.3}$$

To simplify this further, expressing everything in terms of the change in $\mathbf{r}$, we may use the fact that $\dot{\delta\mathbf{r}}=\delta\dot{\mathbf{r}}$, and perform integration by parts to get

$$\delta S = \delta\mathbf{r}\cdot\nabla_{\dot{\mathbf{r}}}L\Big|_{t_i}^{t_f} + \int_{t_i}^{t_f} dt\ \delta\mathbf{r}\cdot\left(\nabla_{\mathbf{r}}L-\frac{d}{dt}\nabla_{\dot{\mathbf{r}}}L\right). \tag{B.1.4}$$

The equations would simplify greatly if the boundary terms vanished. In fact, they do. The reason is that the way the action principle is formulated, the trajectory that makes the action

stationary must respect the boundary conditions of the problem: that we know the position and velocity at the initial and final times. Therefore, if the stationary trajectory respects these boundary conditions, the variation in the trajectory must be zero so that any variation also respects the boundary conditions. Stated mathematically: $\delta\mathbf{r}(t_i) = \delta\mathbf{r}(t_f) = 0$. For

$$0 = \int_{t_i}^{t_f} dt\ \delta\mathbf{r} \cdot \left( \nabla_{\mathbf{r}} L - \frac{d}{dt} \nabla_{\dot{\mathbf{r}}} L \right). \tag{B.1.5}$$

This equation must hold for *any* variation $\delta\mathbf{r}$. The only way this is possible is if the integrand is zero throughout the integration domain. To see this, consider $\delta\mathbf{r} = (1,0,0)\delta(t - t_0)$, with $t_i < t_0 < t_f$. Then we get $0 = \partial_x L(t_0) - \frac{d}{dt}\partial_{\dot{x}} L(t_0)$. This holds for any time, and also for a $y$ or $z$-directed unit vector weighting the delta function. Therefore,

$$\nabla_{\mathbf{r}} L = \frac{d}{dt} \nabla_{\dot{\mathbf{r}}} L, \tag{B.1.6}$$

which is the Euler-Lagrange equation.

To find the Lagrangian of a given system is essentially trial-and-error: guess a Lagrangian and show that the Euler-Lagrange equations result in the equations of motion for the system. While this may sound circular, the purpose of finding a Lagrangian here is to allow us to find the Hamiltonian which lies at the heart of the quantum description of the system. Thus, Lagrangians give a principled way to determine quantized theories of classical systems.

The Lagrangian for a single particle of charge $q$ and mass $m$ in an electromagnetic field can be found to be

$$L = \frac{1}{2} m \dot{\mathbf{r}}^2 - q\phi(\mathbf{r}(t)) + q\mathbf{A}(\mathbf{r}(t), t) \cdot \dot{\mathbf{r}}. \tag{B.1.7}$$

To show this is the correct Lagrangian, let us evaluate the corresponding Euler-Lagrange equation, showing that it reduces to the Newtonian equation of motion for a charged particle subject to the Lorentz force law. The left-hand side of the Euler-Lagrange equation is simply

$$\nabla_{\mathbf{r}} L = -q\nabla\phi + q(\nabla_{\mathbf{r}} \mathbf{A}) \cdot \dot{\mathbf{r}}, \tag{B.1.8}$$

where it is understood that the dot-product refers to the indices of $\mathbf{A}$ and $\mathbf{r}$. The right-hand side of the Euler-Lagrange equation is given by

$$\frac{d}{dt} \nabla_{\dot{\mathbf{r}}} L = m\ddot{\mathbf{r}} + \frac{d}{dt} \mathbf{A}(\mathbf{r}(t), t) \cdot \nabla_{\dot{\mathbf{r}}} \dot{\mathbf{r}} \tag{B.1.9}$$

which, by application of the chain-rule, gives

$$\frac{d}{dt} \nabla_{\dot{\mathbf{r}}} L = m\ddot{\mathbf{r}} + q\partial_t \mathbf{A} + q(\dot{\mathbf{r}} \cdot \nabla)\mathbf{A}, \tag{B.1.10}$$

where we have omitted the functional dependence of $\mathbf{A}$, as we will not need to think about it anymore. We can very clearly see how the electric field arises in this equation, given that $\mathbf{E} = -\nabla\phi - \partial_t \mathbf{A}$. We need to however expose the dependence on the magnetic field. Let us look at the term $q(\nabla_{\mathbf{r}} \mathbf{A}) \cdot \dot{\mathbf{r}}$ in more detail. Writing the $i$-component of this in repeated index notation, we have that

$$q(\nabla_{\mathbf{r}} \mathbf{A}) \cdot \dot{\mathbf{r}} = q(\partial_i A_j - \partial_j A_i)\dot{r}_j + q(\partial_j A_i)\dot{r}_j. \tag{B.1.11}$$

The combination $\partial_i A_j - \partial_j A_i$ appears to be a curl of the vector potential, a magnetic field. To proceed, it is useful to cast the term we want $(\dot{\mathbf{r}} \times (\nabla \times \mathbf{A}))_i$ in a form that resembles what we have just derived. We may write

$$\begin{aligned}(\dot{\mathbf{r}} \times (\nabla \times \mathbf{A}))_i &= \epsilon_{ijk}\dot{r}_j \epsilon_{klm}\partial_l A_m \\ &= \epsilon_{kij}\epsilon_{klm}\dot{r}_j \partial_l A_m \\ &= (\delta_{il}\delta_{jm} - \delta_{im}\delta_{jl})\dot{r}_j \partial_l A_m \\ &= \dot{r}_j(\partial_i A_j - \partial_j A_i),\end{aligned} \tag{B.1.12}$$

exactly equal to a term we got from the Euler-Lagrange equation. Therefore $q(\nabla_{\mathbf{r}}\mathbf{A}) \cdot \dot{\mathbf{r}} = q\dot{\mathbf{r}} \times \mathbf{B} + q(\dot{\mathbf{r}} \cdot \nabla_{\mathbf{r}})\mathbf{A}$. Equating the left and right hand sides of the Euler-Lagrange equations then yields

$$m\ddot{\mathbf{r}} = q(\mathbf{E} + \dot{\mathbf{r}} \times \mathbf{B}), \tag{B.1.13}$$

as desired.

In many cases, we have not just one particle, but many ($N$), in external electromagnetic fields. The arguments of this section can easily be generalized to show that the Euler-Lagrange equations still apply for each particle independently, and that the Lagrangian is the sum of the Lagrangians of each individual particle. In other words

$$L = \sum_{i=1}^{N} \frac{1}{2} m_i \dot{\mathbf{r}}_i^2 - q_i \phi(\mathbf{r}_i, t) + q_i \mathbf{A}(\mathbf{r}_i, t) \cdot \dot{\mathbf{r}}_i \tag{B.1.14}$$

### B.1.2 Lagrangian of the fields without sources

We now move to derive the Lagrangian of the free electromagnetic field. Let us consider for simplicity the electromagnetic field in the absence of sources. Unlike matter, which have well-defined locations, fields are spread out over all space. As a result, the Lagrangian of some field, $\phi$, is typically expressed in terms of a Lagrangian density, $\mathcal{L}$ via

$$L[\phi] = \int d^3r \ \mathcal{L}[\phi(\mathbf{r}, t), \partial_\mu \phi(\mathbf{r}, t)], \tag{B.1.15}$$

where we have taken the field to be in three spatial dimensions, as is the most common scenario. We have assumed the Lagrangian to be a function of both the field itself and its derivatives, labeling a generic derivative (time or space), as $\partial_\mu \phi$. The corresponding action is expressed as

$$S[\phi] = \int d^4x \ \mathcal{L}[\phi(\mathbf{r}, t), \partial_\mu \phi(\mathbf{r}, t)], \tag{B.1.16}$$

with $d^4x \equiv d^3r dt$. Fields, like matter, are also subject to a principle of stationary action. Thus, the field is that which leads to $\delta S = 0$. Let us consider an arbitrary variation of the field $\phi \to \phi + \delta\phi$. The action can then be expanded in this variation to first order as

$$\begin{aligned}S[\phi + \delta\phi] &= \int d^4x \ \mathcal{L}[\phi(\mathbf{r}, t) + \delta\phi, \partial_\mu \phi(\mathbf{r}, t) + \delta(\partial_\mu \phi)] \\ &\approx S[\phi] + \int d^4x \left( \delta\phi \frac{\partial \mathcal{L}}{\partial \phi} + \delta(\partial_t \phi) \frac{\partial \mathcal{L}}{\partial(\partial_t \phi)} + \delta(\nabla_{\mathbf{r}}\phi) \cdot \frac{\partial \mathcal{L}}{\partial(\nabla_{\mathbf{r}}\phi)} \cdot \right),\end{aligned} \tag{B.1.17}$$

where

$$\frac{\partial \mathcal{L}}{\partial(\nabla_{\mathbf{r}}\phi)} \equiv \left(\frac{\partial \mathcal{L}}{\partial(\partial_x\phi)}, \frac{\partial \mathcal{L}}{\partial(\partial_y\phi)}, \frac{\partial \mathcal{L}}{\partial(\partial_z\phi)}\right) \tag{B.1.18}$$

in Cartesian coordinates. To proceed, we need to consider boundary conditions for the fields. Regarding spatial boundary conditions, let us assume that the fields either vanish at infinity, or that the fields are enclosed in a large box with periodic boundary conditions. Similarly, we will apply the same temporal boundary conditions. Whether the fields vanish or are periodic, one can show that the boundary terms in the integration by parts vanish. Therefore, it immediately follows that the variation in the action is

$$\delta S = \int d^4x\ \delta\phi \left(\frac{\partial \mathcal{L}}{\partial \phi} - \partial_t \frac{\partial \mathcal{L}}{\partial(\partial_t\phi)} - \nabla_{\mathbf{r}} \cdot \frac{\partial \mathcal{L}}{\partial(\nabla_{\mathbf{r}}\phi)}.\right) \tag{B.1.19}$$

By an argument similar to the one used to derive the Euler-Lagrange equations for matter, it follows that for the action to be stationary for *any* $\delta\phi$, it must be that

$$\frac{\partial \mathcal{L}}{\partial \phi} - \nabla_{\mathbf{r}} \cdot \frac{\partial \mathcal{L}}{\partial(\nabla_{\mathbf{r}}\phi)} = \partial_t \frac{\partial \mathcal{L}}{\partial(\partial_t\phi)}, \tag{B.1.20}$$

which is the Euler-Lagrange equation for fields.

Let us now apply this to the case of electromagnetism in (non-magnetic, non-dispersive) material media. The Lagrangian of the electromagnetic field, described by a scalar and vector potential, turns out to be

$$L[\phi, \mathbf{A}, \partial_\mu \mathbf{A}] = \frac{\epsilon_0}{2}\int d^3r\ \epsilon\mathbf{E}^2 - c^2\mathbf{B}^2 = \frac{\epsilon_0}{2}\int d^3r\ \epsilon(\nabla\phi + \partial_t\mathbf{A})^2 - c^2(\nabla\times\mathbf{A})^2. \tag{B.1.21}$$

This Lagrangian looks considerably more complicated than the one we derived above for a single-component field due to the dependence on multiple fields $(\phi, \mathbf{A})$, as well as due to the vectorial nature of the vector potential. However, from the standpoint of the derivation of the Euler-Lagrange equation, all that has happened is that the number of degrees of freedom is larger. We can think of the Lagrangian as

$$L[\phi, \partial_\mu\phi, A_x, \partial_\mu A_x, A_y, \partial_\mu A_y, A_z, \partial_\mu A_z]. \tag{B.1.22}$$

And the Euler-Lagrange equations will apply separately to $\phi, A_x, A_y, A_z$.

First, let's find the equation for the scalar potential. Noticing that the Lagrangian does not depend on $\phi$ or $\partial_t\phi$, but does depend on its spatial gradient, we see that the Euler-Lagrange equation generated from the potential is

$$\nabla\cdot\epsilon\nabla\phi + \partial_t\nabla\cdot(\epsilon\mathbf{A}) = 0, \tag{B.1.23}$$

which is in agreement with Eq. (3.1.7) in the absence of sources.

Now, we need to obtain the equations from differentiation with respect to the vector potential and its derivatives. There is no dependence on the vector potential itself, only its derivatives. Differentiating with respect to the time-derivative of $A_s$, the $s$-component of $\mathbf{A}$, gives

$$\frac{\partial \mathcal{L}}{\partial(\partial_t A_s)} = \epsilon_0\epsilon(\partial_t A_s + \partial_s\phi) = -\epsilon_0\epsilon E_s = -D_s, \tag{B.1.24}$$

with $\mathbf{D}$ the displacement field.

Now, we must differentiate the Lagrangian with respect to the spatial derivatives, which is complicated by the presence of curls. The term to evaluate for the equation for $A_s$ is

$$\partial_r \frac{\partial \mathcal{L}}{\partial(\partial_r A_s)}. \tag{B.1.25}$$

In repeated-index notation, the term in the Lagrangian density associated with the magnetic field (modulo $\epsilon_0 c^2$) is

$$-\frac{1}{2}\epsilon_{ijk}\epsilon_{ilm}(\partial_j A_k)(\partial_l A_m). \tag{B.1.26}$$

Noting that the $\partial_i A_j$ are independent degrees of freedom, it follows that $\frac{\partial(\partial_i A_j)}{\partial(\partial_r A_s)} = \delta_{ir}\delta_{js}$. It follows that the derivative (Eq. 5.1.25) of Eq. (5.1.26) is simply

$$\epsilon_{sri}\epsilon_{ilm}\partial_r\partial_l A_m = (\nabla \times \nabla \times \mathbf{A})_s. \tag{B.1.27}$$

Therefore, the equation for the vector potential is

$$\nabla \times \nabla \times \mathbf{A} + \frac{\epsilon}{c^2}\left(\partial_t^2 \mathbf{A} + \nabla \partial_t \phi\right) = 0, \tag{B.1.28}$$

which is Eq. (3.1.8) in the absence of sources. We have thus proven that the Lagrangian of Eq. 5.1.21 is in fact the correct Lagrangian of the field.

### B.1.3 Interaction Lagrangian

Now let's consider the influence of interactions between matter and light. Actually, there's not much to consider. In Section 5.1.1, we derived the Lagrangian describing matter in electromagnetic fields, thus implicitly taking into account the interactions between matter and fields. The only real change is one of perspective: the fields are now degrees of freedom, rather than fixed, external entities, and therefore are subject to time-evolution. The Lagrangian of a system of $N$ matter in an electromagnetic field is thus

$$\boxed{L = \sum_{i=1}^{N} \frac{1}{2} m_i \dot{\mathbf{r}}_i^2 - q_i \phi(\mathbf{r}_i, t) + q_i \mathbf{A}(\mathbf{r}_i, t) \cdot \dot{\mathbf{r}}_i + \frac{\epsilon_0}{2} \int d^3 r \; \epsilon(\nabla\phi + \partial_t \mathbf{A})^2 - c^2(\nabla \times \mathbf{A})^2}. \tag{B.1.29}$$

This Lagrangian does not change the equations of motion for the matter, as the field Lagrangian does not depend on the particle degrees of freedom. However, the term representing the interaction between matter and fields: $-q_i\phi(\mathbf{r}_i, t) + q_i \mathbf{A}(\mathbf{r}_i, t) \cdot \dot{\mathbf{r}}_i$ do change the equation for the fields. To find the change to the field equations however, we need to find the Lagrangian density associated with the interaction terms, and cast these terms as integrals. This is done by introducing integrals, so that the interaction terms become

$$\begin{aligned} &\int d^3 r - q_i \phi(\mathbf{r}_i, t)\delta(r - r_i(t)) + q_i \mathbf{A}(\mathbf{r}_i, t) \cdot \dot{\mathbf{r}}_i \delta(r - r_i(t)) \\ &= \int d^3 r \; - \rho(\mathbf{r}, t)\phi(\mathbf{r}, t) + \mathbf{j}(\mathbf{r}, t) \cdot \mathbf{A}(\mathbf{r}, t), \end{aligned} \tag{B.1.30}$$

where we have introduced the charge and current densities.

Differentiating the Euler-Lagrange equations for the fields gives

$$\nabla \cdot \epsilon \nabla \phi + \partial_t \nabla \cdot (\epsilon \mathbf{A}) = -\rho/\epsilon_0 \tag{B.1.31}$$

for the scalar potential equation and

$$\nabla \times \nabla \times \mathbf{A} + \frac{\epsilon}{c^2}\left(\partial_t^2 \mathbf{A} + \nabla \partial_t \phi\right) = -\mu_0 \mathbf{j} \tag{B.1.32}$$

for the vector potential equation. These are exactly the same as the Maxwell equations with sources that we derived earlier. With this, we have the full *Maxwell-Newton-Lorentz* system of equations that classically describes light and matter

$$\begin{aligned} &m\ddot{\mathbf{r}} = q(\mathbf{E} + \dot{\mathbf{r}} \times \mathbf{B}) \\ &\nabla \cdot \epsilon \nabla \phi + \partial_t \nabla \cdot (\epsilon \mathbf{A}) = -\rho/\epsilon_0 \\ &\nabla \times \nabla \times \mathbf{A} + \frac{\epsilon}{c^2}\left(\partial_t^2 \mathbf{A} + \nabla \partial_t \phi\right) = -\mu_0 \mathbf{j}. \end{aligned} \tag{B.1.33}$$

### B.1.4 Hamiltonian of the matter and the fields

As our ultimate goal is a quantum theory of matter and electromagnetic fields, as because the Hamiltonian is the central quantity of that theory, we must derive the Hamiltonian corresponding to this Lagrangian. As you will remember from mechanics, the Hamiltonian $H(p, q)$ corresponding to a Lagrangian $L(q, \dot{q})$ is obtained by the Legendre transform:

$$H = p\dot{q} - L, \tag{B.1.34}$$

where $p$, the canonical momentum, is defined as

$$p = \frac{\partial L}{\partial \dot{q}}. \tag{B.1.35}$$

For the matter, the canonical momentum can easily be seen to be

$$\mathbf{p} = m\mathbf{v} + q\mathbf{A}. \tag{B.1.36}$$

Meanwhile, for the fields, the canonical momentum is

$$\mathbf{\Pi}(\mathbf{r}) = \frac{\partial L}{\partial \dot{\mathbf{A}}} = -\mathbf{D}. \tag{B.1.37}$$

The Hamiltonian then follows immediately as

$$H = \sum_{i=1}^{N} \frac{(\mathbf{p}_i - q_i \mathbf{A}(\mathbf{r}_i))^2}{2m_i} + q_i \phi(\mathbf{r}_i, t) + \frac{\epsilon_0}{2} \int d^3 r \, \frac{\mathbf{\Pi}^2}{\epsilon} + c^2 (\nabla \times \mathbf{A})^2. \tag{B.1.38}$$

While this answer looks quite clean, there is a serious complication associated with it. It has to do with the fact that matter and field degrees of freedom are mixed together. For example, the vector and scalar potentials in Eq. (5.1.33) are mixed together. Additionally, the

Hamiltonian of the field mixes the scalar and vector potentials through $\Pi$. This complicates analysis greatly, and thus we at this stage make a choice of gauge that eliminates this mixing.

From now on, we will assume to be in the *generalized Coulomb gauge* introduced in Chapter, 3 so that

$$\boxed{\nabla \cdot \epsilon \mathbf{A} = 0}. \tag{B.1.39}$$

This choice of gauge leads to two major simplifications. The first is that the scalar potential is completely determined by the matter (as there are no homogeneous solutions to $\nabla \cdot \epsilon \nabla \phi = -\rho/\epsilon_0$ that carry an electric field). Therefore, the scalar potential is actually a particle degree of freedom. The scalar potential is given in terms of the Green's function of the Laplace equation as

$$\phi(\mathbf{r}) = -\frac{1}{\epsilon_0} \sum_{i=1}^{N} q_i G_\phi(\mathbf{r}, \mathbf{r}_i) \tag{B.1.40}$$

The second thing is that we can define the canonical field momentum without the scalar potential. This is because the canonical field momentum came from deriving a term like

$$\int d^3r \; \epsilon((\partial_t \mathbf{A})^2 + 2\mathbf{A} \cdot \nabla \phi + (\nabla \phi)^2). \tag{B.1.41}$$

But, if $\nabla \cdot \epsilon \mathbf{A} = 0$, then $\int d^3r \; \epsilon \mathbf{A} \cdot \nabla \phi = 0$ because it is an integral of a dot product between a curl-less and divergence-less function. Meanwhile, we can write the third term as

$$\int d^3r \; \epsilon \nabla \phi \cdot \nabla \phi = \int d^3r \; -\phi \nabla \cdot \epsilon \nabla \phi = \int d^3r \; \rho \phi. \tag{B.1.42}$$

Therefore, re-defining the canonical momentum for the field as $\mathbf{\Pi} = \epsilon_0 \epsilon (\partial_t \mathbf{A})$, we can write the Hamiltonian as:

$$H = \sum_{i=1}^{N} \frac{(\mathbf{p}_i - q_i \mathbf{A}_i(\mathbf{r}_i))^2}{2m_i} - \frac{1}{2\epsilon_0} \sum_{i,j=1}^{N} q_i q_j G_\phi(\mathbf{r}_i, \mathbf{r}_j) + \frac{\epsilon_0}{2} \int d^3r \; \frac{\mathbf{\Pi}^2}{\epsilon} + c^2 (\nabla \times \mathbf{A})^2. \tag{B.1.43}$$

This is the final form of the Hamiltonian of light and matter. As a note, while the Hamiltonian "should" be written in terms of position and momenta, as we have above, it is very common to see the Hamiltonian with the field momentum expressed in terms of the vector potential, in order to keep a small number of variables in the Hamiltonian. Thus, the form we will state in practice is

$$\boxed{H = \sum_{i=1}^{N} \frac{(\mathbf{p}_i - q_i \mathbf{A}_i(\mathbf{r}_i))^2}{2m_i} - \frac{1}{2\epsilon_0} \sum_{i,j=1}^{N} q_i q_j G_\phi(\mathbf{r}_i, \mathbf{r}_j) + \frac{\epsilon_0}{2} \int d^3r \; \epsilon (\partial_t \mathbf{A})^2 + c^2 (\nabla \times \mathbf{A})^2}. \tag{B.1.44}$$

### B.1.5 Electromagnetic modes as degrees of freedom of the radiation

The Hamiltonian form of classical electrodynamics exposes that the non-redundant degrees of freedom are the system are the positions and momenta of the matter, the vector potential, and its time-derivative. The vector potential carries an infinite number of degrees of freedom,

as a result of the fact that the fields are defined at every point in space. This is cumbersome, and it would be ideal to have a simpler (but still infinite) set of degrees of freedom by which to describe the electromagnetic field. The mode expansion technique of Chapter 3 provides us with just that.

Recall that: for a time-dependent potential satisfying the generalized Coulomb gauge condition, we may expand the potential in terms of the modes of the source-free Maxwell equations as:

$$\mathbf{A}(\mathbf{r},t) = \sum_n A_n(t)\mathbf{F}_n(\mathbf{r}). \tag{B.1.45}$$

Here, we will take $A_n(t)$ to be purely real coefficients, and $\mathbf{F}_n(\mathbf{r})$ to be real-valued modes. This can be done without loss of generality in lossless non-magnetic systems[1] Later, we will show that this assumption was unnecessary, and that everything we will derive will hold for complex modes as well. But this makes the initial derivations very easy.

Let us see how the Lagrangian and Hamiltonian of the electromagnetic field without sources looks like under this mode expansion. Plugging this into the Lagrangian, and using orthonormality, we immediately find that:

$$L = \frac{\epsilon_0}{2}\sum_n \left(\dot{A}_n^2 - \omega_n^2 A_n^2\right). \tag{B.1.46}$$

The second term arose from moving one of the curls to the other side and making use of the Maxwell eigenproblem. The Lagrangian has now been recast as a function of $A_n$ and $\dot{A}_n$,, suggesting that the mode coefficients, $A_n$, are the canonical coordinates of the electromagnetic field. For each $n$, then we have as the corresponding Euler-Lagrange equation:

$$\ddot{A}_n + \omega_n^2 A_n = 0, \tag{B.1.47}$$

The corresponding Hamiltonian is:

$$H = \frac{\epsilon_0}{2}\sum_n \left(\dot{A}_n^2 + \omega_n^2 A_n^2\right). \tag{B.1.48}$$

Expressing $H$ in terms of the canonical position $A_n$ and momentum $\epsilon_0\dot{A}_n$, we have that

$$H = \sum_n \left(\frac{p_n^2}{2\epsilon_0} + \frac{1}{2}\epsilon_0\omega_n^2 q_n^2\right). \tag{B.1.49}$$

This is clearly the Hamiltonian of a set of harmonic oscillators associated with each mode. The frequencies of the oscillators are the modal frequencies $\omega_n$ and a "mass" $\epsilon_0$.

The result we arrived at, although very simple, is extremely important. It says: **The time-dependent amplitudes of the modes behave in exactly the same way as a harmonic oscillator. In the quantum theory, the mode amplitudes act as independent quantum harmonic oscillators.**

[1] It is a consequence of $\epsilon$ being real that all of the modes can be chosen *real*. It can be immediately seen that if $\mathbf{F}$ is a mode of Maxwell's equations with frequency $\omega$, it is also the case that $\mathbf{F}^*$ is also a mode with the same frequency, meaning that $\mathbf{F} \pm \mathbf{F}^*$ are both modes, which are purely real. This also means that for a non-degenerate mode, it is real. While for a complex mode, it must have a degenerate partner which is its conjugate. This situation also happens in the Schrodinger equation.

This point holds not just for electromagnetism, but other wave equations, such as acoustic wave equations, hydrodynamic wave equations, and relativistic wave equations for fundamental particles. In fact, the statement in the box is the fundamental principle of quantum field theory.

# Appendix C

# Time-evolution in quantum mechanics

## C.1 Time-evolution of quantum systems

In order to describe the quantum interactions of light and matter, we will begin by reviewing the fundamental mathematics which govern the evolution of general quantum systems. Then, we will develop a framework of perturbation theory which is most useful for our purposes.

A highly general quantum mechanical system with Hamiltonian $H(t)$ can be described as the unitary time-evolution of a state vector $|\psi(t)\rangle$ via the Schrödinger equation

$$i\hbar\partial_t |\psi(t)\rangle = H(t) |\psi(t)\rangle . \tag{C.1.1}$$

We can then define the unitary time-evolution operator $U(t, t')$ to express the time-dependent state $|\psi(t)\rangle$ in terms of the state at a previous initial time $t_{init}$ as

$$|\psi(t)\rangle = U(t, t_{init}) |\psi(t_{init})\rangle . \tag{C.1.2}$$

As a brief reminder, the unitary time evolution operator satisfies a few simple but important properties:

1. $U(t_1, t_1) = 1$, where 1 is the identity operator – letting no time pass will not evolve the state
2. $U(t_3, t_2)U(t_2, t_1) = U(t_3, t_1)$ – sequential time evolution
3. $U(t_1, t_2) = U^\dagger(t_1, t_2)$ – unitarity.

The time-dependent Schrodinger equation can then be equivalently formulated as an equation for the unitary time-evolution operator,

$$i\hbar\partial_t U(t, t_{init}) = H(t)U(t, t_{init}). \tag{C.1.3}$$

This has the benefit that it removes reference to the time dependent state $|\psi(t)\rangle$, allowing us to work with operators.

We will now "solve" this equation in a self-consistent manner. We begin by formally integrating both sides of the operator equation:

$$i\hbar \int_{t_{init}}^{t} dt' \; \partial_t U(t', t_{init}) = i\hbar(U(t, t_{init}) - U(t_{init}, t_{init}))$$
$$= \int_{t_{init}}^{t} dt' \; H(t')U(t', t_{init}). \tag{C.1.4}$$

Using property 1 from above, we may then write

$$U(t, t_{init}) = 1 + \left(\frac{1}{i\hbar}\right) \int_{t_{init}}^{t} dt' \; H(t')U(t', t_{init}). \tag{C.1.5}$$

We have converted a differential equation into an integral equation, since $U$ is expressed in terms of itself through an integral. Although this will not provide us with an analytical solution, it will provide a form useful for further theoretical analysis. To derive this form, we plug the left-hand side of the equation into the right-hand side, which gives

$$U(t, t_{init}) = 1 + \left(\frac{1}{i\hbar}\right) \int_{t_{init}}^{t} dt' \; H(t')$$
$$+ \left(\frac{1}{i\hbar}\right)^2 \int_{t_{init}}^{t} dt' \int_{t_{init}}^{t'} dt'' \; H(t')H(t'')U(t'', t_{init}). \tag{C.1.6}$$

Doing this *ad infinitum*, and relabeling the times by numerical indices $(t_1, t_2, \ldots)$, we immediately have

$$U(t, t_{init}) = \sum_{n=0}^{\infty} \left( \prod_{i=1}^{n} \left(\frac{1}{i\hbar}\right) \int_{t_{init}}^{t_{i-1}} dt_i \; H(t_i) \right), \tag{C.1.7}$$

where $t_0 \equiv t$. We now have $U$ in terms of an infinite series which depends only on the Hamiltonian which we know. Eventually, we will invoke perturbation theory to show that we can learn an incredible amount even from the first few terms. To work toward this goal, we can do a few things to make this expression even nicer. It would be convenient if all of the upper limits were the same, so that manipulating the integrals was less cumbersome. This can always be done by extending the limits of integration, and then introducing heaviside step functions into the integrand to restrict the integration range. Rather then going into it in great detail, we will just state the final result, and we encourage you to check that it is equivalent to the previous formulation. Consider a term

$$\int_{t_{init}}^{t} dt_1 \cdots \int_{t_{init}}^{t_{n-1}} dt_n \; H(t_1) \cdots H(t_n). \tag{C.1.8}$$

We claim that it is equal to

$$\frac{1}{n!}\int_{t_{init}}^{t} dt_1 \cdots \int_{t_{init}}^{t_{n-1}} dt_n \mathrm{T}\left[H(t_1)\cdots H(t_n)\right], \tag{C.1.9}$$

where T is the time-ordering operator, which is defined such that "operators at earlier times go to the right of operators at later times." For example, if $A(t)$ is some operator, and $t_2 > t_3 > t_1$, then the action of the time ordering operator is

$$\mathrm{T}\left[A_1(t_1)A_2(t_2)A_3(t_3)\right] = A_2(t_2)A_3(t_3)A_1(t_1). \tag{C.1.10}$$

In order to avoid thinking about case work for all possible orderings of times, we can instead use the heaviside function $\theta(t)$ to enumerate these cases as

$$\begin{aligned}\mathrm{T}\left[A_1(t_1)A_2(t_2)A_3(t_3)\right] &= A_1(t_1)A_2(t_2)A_3(t_3)\theta(t_1-t_2)\theta(t_2-t_3)\\ &\quad + \text{permutations of 1,2,3.}\end{aligned} \tag{C.1.11}$$

From here, one can use this type of reasoning to compactly write the series expansion for the unitary evolution operator as

$$\boxed{U(t,t_{init}) = \mathrm{T}\sum_{n=0}\frac{1}{n!}\left(\frac{1}{i\hbar}\int_{t_{init}}^{t} dt'\ H(t')\right)^n = \mathrm{T}\ \exp\left[-\frac{i}{\hbar}\int_{t_{init}}^{t} dt'\ H(t')\right].} \tag{C.1.12}$$

The right-hand side is called the *time-ordered exponential*. It is important to note that so far, we have made *no approximations*. In the formal sense, this series solution for the time evolution operator is exact. This series representation of the time-evolution operator is a simple result with a myriad of useful applications, and is applied throughout almost the entirety of the text.

**Simplifications of the time-ordered exponential**

Before applying this expression to derive perturbation theory, we should briefly mention two special cases of this expression. First, consider the case in which the Hamiltonian *commutes with itself at later times*: $[H(t), H(t')] = 0$ for all $t, t'$ (this is not common for time-dependent Hamiltonians). In that case, it follows that the time-ordering is ineffectual: all orderings of the Hamiltonian operators are exactly the same. Another way to say this is that the time-ordering operator is the identity, and can thus be ignored. The unitary time-evolution operator is then

$$U(t,t_{init}) = \exp\left[-\frac{i}{\hbar}\int_{t_{init}}^{t} dt'\ H(t')\right]. \tag{C.1.13}$$

The second important case is that in which the Hamiltonian is *time-independent*. Then, the time-ordered exponential reduces to the well-known expression

$$U(t,t_{init}) = \exp\left[-\frac{iH(t-t_{init})}{\hbar}\right]. \tag{C.1.14}$$

**The interaction picture**

We should mention yet another useful representation of the results developed above. Suppose we express the Hamiltonian in the form $H = H_0 + V$, where $H_0$ is some Hamiltonian for which we know the corresponding unitary time-evolution operator $U_0$, and $V$ is a Hamiltonian we know less about. This situation comes up centrally in *perturbation theory*, which we take up in the next section. We may then write the time-dependent Schrodinger equation for the unitary time-evolution operator as

$$i\hbar\partial_t U = (H_0 + V)U, \tag{C.1.15}$$

where we now omit the time-labels, leaving them implicit. Now, let us factor the evolution operator as $U = U_0 U_I$, where $i\hbar\partial_t U_0 = H_0 U_0$, and $U_I$ will be called the *interaction picture time-evolution operator*. It follows immediately that

$$i\hbar\partial_t U_I = U_0^\dagger V U_0 U_I \equiv V_I U_I, \tag{C.1.16}$$

where $V_I$ is the $V$ operator in the so-called *interaction picture*. It should be clear that because this equation is the same as the one we started with in the beginning of this section, the solution is:

$$U_I = \mathrm{T}\exp\left[-\frac{i}{\hbar}\int dt'\ V_I(t')\right]. \tag{C.1.17}$$

With these fundamentals established, we can move on to develop a key analytical tool of quantum mechanics: *time-dependent perturbation theory*.

### C.1.1 Time-dependent perturbation theory

The quantum dynamics of most Hamiltonians cannot be solved for anlytically. Therefore, we must turn to approximate techniques to study the dynamics of quantum systems. An extremely powerful and versatile technique to approximate the dynamics of quantum systems is time-dependent perturbation theory, which allows us to solve for the time-dynamics of a system approximately, provided that the Hamiltonian can be decomposed into the sum of an exactly solvable part plus a small correction. While this may sound artificial, it is fortunately not, and perturbation theory is perhaps one of the most useful tools in quantum mechanics. The premise of perturbation theory is simple, and the time-ordered exponential has perfectly set the stage for this development. We learned from the last section that the unitary time-evolution operator for a general quantum system with Hamiltonian $H = H_0 + V$ can be written as

$$U(t,t_0) = U_0(t,t_0)U_I(t,t_0) = U_0(t,t_0) \times \mathrm{T}\ \exp\left[-\frac{i}{\hbar}\int_{t_0}^{t} dt'\ V_I(t')\right]. \tag{C.1.18}$$

We will often call $H_0, U_0$, and the associated eigenstates of $H_0$ *unperturbed* quantities. The utility of the interaction picture is that it isolates the part of the Hamiltonian which we still need to understand the effects of. The associated probability that the system is in some eigenstate $|f\rangle$ of the unperturbed system at time $t$ is then simply

$$|\,\langle f|U(t,t_0)|i\rangle\,|^2 = |\,\langle f|U_I(t,t_0)|i\rangle\,|^2, \tag{C.1.19}$$

since the action of $U_0$ on $|f\rangle$ is simply to produce a phase. In many problems in perturbation theory, we are interested in *scattering probabilities*, which tell us the probability that a system in state $|i\rangle$ has changed, or "scattered", into state $|f\rangle$. We quickly note an important additional piece of terminology, which is that $U_I$ is often called the "$S$ matrix" or "scattering matrix", and its matrix elements are called $S$-matrix elements. We will use this terminology almost exclusively.

For physical context, we list a few examples of processes which can be described this way

- An excited atom or molecule relaxes to a lower energy level, producing a photon
- A photon incident on an atom or molecule is scattered into a different direction
- An electron in a semiconductor absorbs a photon, exciting the electron from the valence band into the conduction band

We also often formulate this problem in terms of *transition rates*, which tells us the rate at which these scattering probabilities change. In other words, we consider some physical process defined by initial and final states, and then compute the rate at which this process occurs.

The program of perturbation theory for computing these rates is then simple: we Taylor-expand the time-ordered exponential, and find transition rates at whatever order is needed to accurately describe them. In practice, orders higher than two (counting from zero) are rarely considered, as most processes happen to leading order at first or second order in the Taylor expansion. And if higher orders are still needed, it is either for extremely precise predictions (in excess of 8 decimal places of accuracy, which is outside of our scope) or a different method should be found.

**First-order scattering amplitude and transition rate**

First, we discuss first-order perturbation theory – in other words, processes described by the first order term in the time-ordered exponential expansion. Consider a system starting in an eigenstate $|i\rangle$ of the unperturbed system. At first-order in the Taylor expansion, the S-matrix element describing the state $|i\rangle$ scattering into an orthogonal final state $|f\rangle$ is given by

$$S_{fi} = -\frac{i}{\hbar}\int_{-T}^{T} dt'\ \langle f|V_I(t')|i\rangle = -\frac{iV_{fi}}{\hbar}\int_{-T}^{T} dt'\ e^{i\omega_{fi}t} = -\frac{iV_{fi}}{\hbar}\left(2T\mathrm{sinc}\left(\omega_{fi}T\right)\right). \tag{C.1.20}$$

Regarding the limits of integration, we have simply redefined $t$ such that $t - t_0 = 2T$, where $2T$ is *the interaction time interval*, and shifted the origin of time so that this interval is centered around time zero. Recall that the sinc function $\mathrm{sinc}(\alpha x) \equiv \frac{sin(\alpha x)}{\alpha x}$ is extremely sharply peaked around $x = 0$ for large $\alpha$ (and $\mathrm{sinc}(0) = 1$). For very large times then, the only probabilities that are significant are energy-conserving ones: $\omega_f = \omega_i$. Another point to mention is that $\frac{\alpha}{\pi}\int_{-\infty}^{\infty} dx\ \mathrm{sinc}(\alpha x) = 1$. Therefore, in the limit of long times, which is of most interest $T \gg \frac{1}{\omega_{fi}}$, we may say that $2T\mathrm{sinc}(\omega_{fi}T) \to 2\pi\delta(\omega_{fi})$.

Squaring the $S$-matrix to obtain a probability, we have that the transition probability, $P_{fi}$ satisfies:

$$P_{fi} \equiv |S_{fi}|^2 = \frac{|V_{fi}|^2}{\hbar^2}(2\pi)^2\delta^2(\omega_{fi}) = \frac{|V_{fi}|^2}{\hbar^2}(2\pi)^2\delta(\omega_{fi})\delta(0), \tag{C.1.21}$$

where we have used the delta function identity $\delta(x-a)\delta(f(x)-b) = \delta(x-a)\delta(f(a)-b)$, where $f(x) = x$. If you've never seen $\delta(0)$ before, don't freak out. Remember how we got the delta function: from the integral of an exponential over *finite* limits of integration. In other words, $\delta(0)$ should be understood as

$$\delta(0) \to \int_{-T}^{T} \frac{dt}{2\pi} e^{i\omega_{fi}0} = \frac{2T}{2\pi}. \tag{C.1.22}$$

You should not be too stressed about the fact that we used a delta-function identity for something that wasn't really a delta function due to finite limits of integration. That identity generally sharply peaked functions. We will do these kinds of manipulations often, so we encourage you to try this with the sinc functions directly and convince yourself it works out the same way in the desired limit.

With those disclaimers out of the way, the transition probability is given by

$$P_{fi} = (2T)\frac{2\pi}{\hbar^2}|V_{fi}|^2\delta(\omega_{fi}). \tag{C.1.23}$$

The transition probability is directly proportional to the interaction time, $2T$. We define the average transition rate as $\Gamma_{fi} = \frac{P_{fi}}{2T}$. Finally, we are often not only interested in the rate of transitions into one final states, but all possible final states, denoted $\Gamma_i$. This is often called the *decay rate* or *transition rate* of the state $i$. We note that it is conventional to drop the $i$ subscript, as it can be inferred from context which state is the initial state from the matrix element. Noting that $\Gamma = \sum_f \Gamma_{fi}$, we have that

$$\boxed{\Gamma = \frac{2\pi}{\hbar^2}\sum_f |V_{fi}|^2\delta(\omega_{fi}).} \tag{C.1.24}$$

This formula is called *Fermi's golden rule*, and is perhaps one of the most used formulae in quantum mechanics. It is a workhorse formula in atomic and optical physics, condensed matter physics, quantum field theory, and many other areas that rely on quantum mechanics.

Before moving on to second-order scattering probabilities and transition rates, we should quickly mention a corollary of the considerations of this section. What if our initial state is not prepared in an eigenstate of the Hamiltonian, but is instead prepared in a superposition of eigenstates $|i\rangle = \sum_k c_k |k\rangle$ ? The associated scattering probability to a final state $f$ is then given by:

$$S_{fi} = \sum_k c_k S_{fk}. \tag{C.1.25}$$

As probabilities are the squares of these quantities, it is immediately clear that there is a possibility for different terms to interfere via cross-terms.

### Second-order scattering amplitude and transition rates

A number of important physical processes are described at leading order in second-order perturbation theory. Examples of such processes include the scattering of light by electrons

(Rayleigh, Thomson, and Compton scattering), as well as two-photon absorption and emission processes, which are fundamental processes of nonlinear optics. The evaluation of a scattering probability in second-order perturbation theory is in principle a straightforward extension of the considerations of the previous section: we simply expand the unitary evolution operator $U_I$ to second-order. There are a few new steps which are involved, so we outline them here.

The S-matrix element $S_{fi}$, assuming an initial eigenstate $|i\rangle$, and a final eigenstate $|f\rangle$, of the unperturbed Hamiltonian, is given by

$$S_{fi}^{(2)} = \frac{1}{2}\left(-\frac{i}{\hbar}\right)^2 \int_{-T}^{T} dt'dt''\ \langle f|\mathrm{T}\left[V_I(t')V_I(t'')\right]|i\rangle. \tag{C.1.26}$$

The time-ordering can be expressed equivalently (by virtue of its definition) as

$$S_{fi}^{(2)} = \frac{1}{2}\left(-\frac{i}{\hbar}\right)^2 \int_{-T}^{T} dt'dt''\ \langle f|\theta(t'-t'')V_I(t')V_I(t'') + \theta(t''-t')V_I(t'')V_I(t')|i\rangle. \tag{C.1.27}$$

We focus on the first of the two terms, as the second is evaluated in the same way. To put the expression into a convenient form, it is useful to insert a complete set of eigenstates of the unperturbed Hamiltonian, so that we instead evaluate

$$\frac{1}{2}\sum_n \left(-\frac{i}{\hbar}\right)^2 \int_{-T}^{T} dt'dt''\ \theta(t'-t'')\langle f|V_I(t')|n\rangle\langle n|V_I(t'')|i\rangle. \tag{C.1.28}$$

If the unperturbed Hamiltonian is time-independent, the time-dependence of the matrix element reduces the previous expression to

$$\frac{1}{2}\sum_n \left(-\frac{i}{\hbar}\right)^2 \int_{-T}^{T} dt'dt''\ \theta(t'-t'')V_{fn}V_{ni}e^{i\omega_{fn}t'+i\omega_{ni}t''}. \tag{C.1.29}$$

We see that the integral is something like a Fourier transform of the Heaviside step function. This integral may be evaluated by means of a famous formula which is the contour integral representation of the step function.

$$\theta(t'-t'') = -\lim_{\eta\to 0}\int \frac{d\omega}{2\pi i}\ \frac{e^{-i\omega(t'-t'')}}{\omega+i\eta}. \tag{C.1.30}$$

Plugging this formula in, and extending the limits of integration from $-\infty$ to $\infty$ yields

$$i\pi\sum_n \left(-\frac{i}{\hbar}\right)^2 \frac{V_{fn}V_{ni}}{\omega_{in}+i\eta}\delta(\omega_{fi}). \tag{C.1.31}$$

It is simple to verify that the second term in the time-ordering is identical. Therefore, the S-matrix element at second order is given by

$$S_{fi}^{(2)} = 2\pi i\left(-\frac{i}{\hbar}\right)^2 \sum_n \frac{V_{fn}V_{ni}}{\omega_{in}+i\eta}\delta(\omega_{fi}). \tag{C.1.32}$$

Squaring the S-matrix element to obtain a transition probability, handling $\delta(0)$ the same way as in the previous section, and summing over final states, immediately yields Fermi's Golden Rule at second order:

$$\boxed{\Gamma = \frac{2\pi}{\hbar^4} \sum_f \left| \sum_n \frac{V_{fn} V_{ni}}{\omega_{in} + i\eta} \right|^2 \delta(\omega_{fi}).} \tag{C.1.33}$$

The limit $\eta \to 0$ is implicit. You might wonder why the denominator is being made slightly complex. It is to handle an important case of second-order perturbation theory: resonance, i.e., when for some *intermediate state* or *virtual state* $n$, $\omega_{ni} = 0$ (and by energy conservation $\omega_{fn} = 0$). We will not consider this case here. We will simply content ourselves by mentioning that when resonance happens, the decay rate is essentially that of two sequential first-order processes: from $i$ to $n$ at first order, and then from $n$ to $f$ at first order. This is important in the consideration of elementary physical processes such as resonance flourescence and radiative cascade.

**Energy shifts in time-dependent perturbation theory**

In the previous two sections, we looked at S-matrix elements between different final states. This leads to a natural question: what about when the two states are the same? It turns out that these elements of the S-matrix lead to energy shifts. You may remember from an elementary quantum mechanics course that energy shifts are typically evaluated in time-independent perturbation theory. We will now show how to evaluate them in time-dependent perturbation theory, and that this yields the same results as from time-independent perturbation theory. The main advantage of evaluating energy shifts in time-dependent perturbation theory is economy of formalism: we can now use a single method (time-dependent perturbation theory) to evaluate all perturbation theory quantities of interest.

To see that elements like $S_{ii}$ are related to energy shifts, consider $S_{ii}(T, -T)$ up to second-order in perturbation theory, a la Equations (15) and (27). Restoring the zeroth order contribution, we can write

$$S_{ii}(T, -T) \approx 1 - \frac{iV_{ii}}{\hbar}(2T) - \frac{i}{\hbar} \sum_n \frac{V_{in} V_{ni}}{\hbar\omega_{in} + i\hbar\eta}(2T) \equiv 1 - \frac{i\delta E(2T)}{\hbar}, \tag{C.1.34}$$

where we define an energy shift $\delta E$ as

$$\delta E = V_{ii} + \sum_n \frac{V_{in} V_{ni}}{\hbar\omega_{in} + i\hbar\eta}. \tag{C.1.35}$$

Noting that for small changes in energy, $1 - \frac{i\delta E(2T)}{\hbar} \approx e^{-\frac{i\delta E(2T)}{\hbar}}$, and recalling that the S-matrix represents time-evolution *in addition* to the unperturbed Hamiltonian, we see that in that case, the evolution of the initial state can then be approximated by $e^{-\frac{i(E_i + \delta E)(2T)}{\hbar}}$. We may then claim that the effect of the perturbation is to shift the energy of the state $|i\rangle$ by an amount given by Equation (35). You may recognize that that energy shift is exactly the one derived through non-degenerate time-independent perturbation theory. There is one additionally interesting thing to note. In the second-order contribution to the energy shift, we again see the possibility of resonance for some intermediate state $|f\rangle$. Evaluating its contribution to

the energy shift by contour integration will yield a complex energy, whose imaginary part is given by (half) the decay rate given by Fermi's Golden Rule at first order in perturbation theory. In other words, decay leads to "complex energies", very roughly speaking.

**Eigenfunction shifts in time-dependent perturbation theory**

You may remember that in time-independent perturbation theory, shifts in the eigenfunctions can also be calculated. We can also do it in time-dependent perturbation theory. Consider a state which starts at time $-T$ in an eigenstate $|n\rangle$ of $H_0$ and a perturbation $V$ is turned on. We will assume there is no degeneracy between discrete states. The resulting state at first order is

$$|\psi(t)\rangle = U_0(t,t_0)\left(1+\frac{1}{i\hbar}\int_{-T}^{t} dt' V_I(t')\right)|n\rangle \tag{C.1.36}$$

Expressing $V_I = U_0^\dagger V U_0$, and inserting a complete set of states, we have that

$$|\psi(t)\rangle = e^{-i\omega_n t}|n\rangle + \frac{1}{i\hbar}U_0(t,t_0)\int_{-T}^{t}\sum_m e^{i\omega_{mn}t}V_{mn}|m\rangle. \tag{C.1.37}$$

Regulating the interaction by turning it on slowly with a time-dependence $e^{-\eta|t|}$, with $\eta$ infinitesimal, we have:

$$|\psi(t)\rangle = e^{-i\omega_n(t+T)}\left(1-\frac{iV_{nn}}{\hbar}(t+T)\right)|n\rangle - e^{i\omega_m(t+T)}\sum_{m\neq n}\frac{V_{mn}}{\hbar\omega_{mn}}|m\rangle. \tag{C.1.38}$$

Where wehave dropped the complex infinitesimal in the denominator on the assumption of non-degeneracy. The third term follows from the considerations of the previous section. Approximating the terms in parentheses as an exponential as in the previous subsection, we have

$$|\psi(t)\rangle = e^{-i\left(\omega_n+\frac{V_{nn}}{\hbar}\right)(t+T)}|n\rangle + e^{i\omega_m(t+T)}\sum_{m\neq n}\frac{V_{mn}}{\hbar\omega_{nm}}|m\rangle. \tag{C.1.39}$$

The first term is the initial state with the first-order energy shift. The second term is the static eigenfunction shift from first-order time-independent perturbation theory.

### C.1.2 Worked examples of transition rates

In this section, we will do a couple of examples of transition rate problems, which will (a) give you a clearer idea of how the formulae developed here are applied, (b) be of great relevance later in the text, and (c) impress upon you that some very important phenomena can be explained as decays.

**Quantum theory of friction (two-level system coupled to harmonic oscillators)**

Let's consider a two-level system which is coupled to many harmonic oscillators all of mass $m$. This problem comes up a lot in the quantum theory of damping (friction), as well as in atomic

physics, when considering spontaneous emission by atoms. Let us assume that the two-level system has an energy difference between the excited and ground states of $\hbar\omega_0$. Let us also assume that the oscillators are labeled by a one-dimensional continuous index $k$, and that the oscillators have a continuum of frequencies $\omega_k$. Let us further assume that the interaction is between the $\sigma_x$ operator of the two-level system and the position operator of each of the oscillators, with an overall coupling scale $\alpha_j$, such that the Hamiltonian for this system is:

$$H = \frac{1}{2}\hbar\omega_0\sigma_z + \sum_k \hbar\omega_k \left(a_k^\dagger a_k + \frac{1}{2}\right) + \sum_k \alpha_j \sqrt{\frac{\hbar}{2m\omega_k}}(a_k + a_k^\dagger)\sigma_x. \tag{C.1.40}$$

We may readily observe that the first two terms of the Hamiltonian correspond to a system which we may readily diagonalize. The eigenstates are of the form

$$|g\rangle|n_1 n_2 \cdots\rangle, |e\rangle|n_1 n_2 \cdots\rangle, \tag{C.1.41}$$

with corresponding energies

$$-\frac{1}{2}\hbar\omega_0 + \sum_k \hbar\omega_k \left(n_k + \frac{1}{2}\right), \frac{1}{2}\hbar\omega_0 + \sum_k \hbar\omega_k \left(n_k + \frac{1}{2}\right) \tag{C.1.42}$$

Note that as the zero of energy may be shifted arbitrarily, we can shift every state down by $\sum_k \frac{1}{2}\hbar\omega_k$, even though for a continuum, this is infinite. We thus identify the first two terms of the Hamiltonian as $H_0$, the unperturbed Hamiltonian. The perturbation, $V$, is simply the third term. It's effect vanishes when $\alpha_j = 0$ for all $j$.

Suppose the initial state of the system at time 0 is $|e\rangle|0\cdots 0\rangle$. In other words, the two-level system is in the excited state, and the harmonic oscillators are all in the ground state. To determine possible decay pathways at first order, we ask: what states can the interaction Hamiltonian take the initial state to. Apply $V$ to this state. $\sigma_x$ flips $|e\rangle$ to $|g\rangle$. The sum of creation and annihilation operators promotes the vacuum of oscillators to a superposition of states where each state individually has an excitaton of the oscillator. Therefore, a suitable basis of final states is of the form

$$|f\rangle = |g\rangle|0_1 \cdots 0_{j-1} 1_j 0_{j-1} \cdots\rangle. \tag{C.1.43}$$

Simply transcribing equation (24), and evaluating the matrix elements of the harmonic oscillator, we see that the transition rate is

$$\Gamma = \frac{2\pi}{\hbar}\sum_j \frac{\alpha_j^2}{2m\omega_j}\delta(\omega_{eg} - \omega_j). \tag{C.1.44}$$

The continuous sum can also be written as an integral using the notion of *density of states*. The density of states $\rho(\omega) = \frac{dN}{d\omega}$ represents the number of states $dN$ in an interval of energies $\hbar d\omega$ centered around energy $\hbar\omega$. It's integral represents the total number of states. The decay rate may then be written as

$$\Gamma = \frac{2\pi}{\hbar}\rho(\omega_0)\frac{\alpha^2(\omega_0)}{2m\omega_0}. \tag{C.1.45}$$