Unit 5: Quantum effects in nonlinear optics

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Contents

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.

I. 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 ω_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 ω_1 or ω_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 was also the case classically). To do this, we introduce

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[1] We stated this was the photon picture of parametric down conversion in Unit 2, but could not prove this, of course.

the so-called positive and negative frequency parts of the electric field operator:

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}^{(+)}(\mathbf{r},t) + \mathbf{E}^{(-)}(\mathbf{r},t). \tag{I.1}$$

The positive and negative frequency parts of the field operator are defined such that

$$\mathbf{E}^{(\pm)}(\mathbf{r},t) = \int_0^\infty \frac{d\omega}{2\pi} e^{\mp i\omega t} E^{(\pm)}(\mathbf{r},\omega). \tag{I.2}$$

Let us now consider for example the Fourier component of the electric field operator at ω (associated with $e^{-i\omega t}$). It follows from these definitions, and the operator Maxwell equations, that:

$$\nabla \times \nabla \times \mathbf{E}^{(+)}(\mathbf{r},\omega) - \epsilon(\omega)\frac{\omega^2}{c^2}\mathbf{E}^{(+)}(\mathbf{r},\omega) = \mu_0\omega^2\mathbf{P}^{(+)}(\mathbf{r},\omega), \tag{I.3}$$

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

$$-\left(\nabla^2 + \epsilon(\omega)\frac{\omega^2}{c^2}\right)\mathbf{E}^{(+)}(\mathbf{r},\omega) = \mu_0\omega^2\mathbf{P}^{(+)}(\mathbf{r},\omega). \tag{I.4}$$

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

$$\mathbf{E}^{(+)}(\mathbf{r},\omega) = \mathbf{E}_1^{(+)}(\mathbf{r},\omega) + \mathbf{E}_2^{(+)}(\mathbf{r},\omega), \tag{I.5}$$

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

$$\mathbf{E}^{(+)}(\mathbf{k}_\perp, z, \omega) = \mathbf{E}_1^{(+)}(\mathbf{k}_\perp, z, \omega) + \mathbf{E}_2^{(+)}(\mathbf{k}_\perp, z, \omega). \tag{I.6}$$

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

$$E^{(+)}(\mathbf{k}_\perp, z, \omega) = E_1^{(+)}(\mathbf{k}_\perp, z, \omega)\hat{\epsilon}_{1,\mathbf{k}} + E_2^{(+)}(\mathbf{k}_\perp, z, \omega)\hat{\epsilon}_{2,\mathbf{k}}, \tag{I.7}$$

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

$$-\left(\frac{d^2}{dz^2} + \left(\frac{n_{\text{eff}}(\omega)\omega}{c}\right)^2 - k_\perp^2\right) E_1^{(+)}(\mathbf{k}_\perp, z, \omega) = \mu_0\omega^2\hat{\epsilon}_{1,\mathbf{k}} \cdot \mathbf{P}_1^{(+)}(\mathbf{k}_\perp, z, \omega), \tag{I.8}$$

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

$$-\left(\frac{d^2}{dz^2} + k_{1,z}^2(\omega)\right) E_1^{(+)}(\mathbf{k}_\perp, z, \omega) = \mu_0\omega^2\hat{\epsilon}_{1,\mathbf{k}} \cdot \mathbf{P}_1^{(+)}(\mathbf{k}_\perp, z, \omega), \tag{I.9}$$

We now make a slowly varying envelope approximation. We write

$$E_1^{(+)}(\mathbf{k}_\perp, z, \omega) = A_1^{(+)}(\mathbf{k}_\perp, z, \omega)e^{ik_{1,z}(\omega_1)z}, \tag{I.10}$$

and neglect terms associated with the second derivative of the slow operator. This turns the left-hand side into

$$-\left(2ik_{1,z}(\omega_1)\partial_z A_1^{(+)}(\mathbf{k}_\perp, z, \omega) + \left(k_{1,z}^2(\omega) - k_{1,z}^2(\omega_1)\right) A_1^{(+)}(\mathbf{k}_\perp, z, \omega)\right). \tag{I.11}$$

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 ω_1, writing

$$k_{1,z}(\omega) = k_{1,z}(\omega_1) + \sum_{m=1}^{\infty} \frac{\beta_m}{m!}(\omega - \omega_1)^m. \tag{I.12}$$

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

$$-\left(2ik_{1,z}(\omega_1)\partial_z \tilde{A}_1^{(+)}(\mathbf{k}_\perp, z, t) + 2k_{1,z}(\omega_1)\sum_{m=1}^{\infty}\frac{i^m\beta_m}{m!}\partial_t^m \tilde{A}_1^{(+)}(\mathbf{k}_\perp, z, t)\right). \tag{I.13}$$

The polarization terms in the time-domain may be written as (in envelope form)

$$\mu_0\omega_1^2\hat{\epsilon}_{1,\mathbf{k}}\cdot\tilde{\mathbf{P}}_1^{(+)}(\mathbf{k}_\perp, z, t), \tag{I.14}$$

and so putting everything together, we have

$$\partial_z \tilde{A}_1^{(+)}(\mathbf{k}_\perp, z, t) = \sum_{m=1}^{\infty}\frac{i^{m+1}\beta_m}{m!}\partial_t^m \tilde{A}_1^{(+)}(\mathbf{k}_\perp, z, t) + \frac{i\omega_1^2}{2\epsilon_0 c^2 k_{1,z}(\omega_1)}\hat{\epsilon}_{1,\mathbf{k}}\cdot\tilde{\mathbf{P}}_1^{(+)}(\mathbf{k}_\perp, z, t). \tag{I.15}$$

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!

II. 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

$$\partial_z \tilde{A}_1^{(+)}(\mathbf{k}_{1,\perp}, z, t) = \frac{i\omega_1^2}{2\epsilon_0 c^2 k_{1,z}(\omega_1)} \hat{\epsilon}_{1,\mathbf{k}} \cdot \tilde{\mathbf{P}}_1^{(+)}(\mathbf{k}_{1,\perp}, z, t), \tag{II.1}$$

where I have taken $\mathbf{k}_\perp \to \mathbf{k}_{1,\perp}$ in order to remind us that we're taking about the ω_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]

$$\mathbf{E}^{(+)}(\mathbf{r}, t) \to \left(A_3 e^{i\mathbf{k}_3\cdot\mathbf{r} - i\omega_3 t}\hat{\epsilon}_3 + \text{c.c.}\right) + \mathbf{E}^{(+)}(\mathbf{r}, t), \tag{II.2}$$

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

$$P_i(\mathbf{r}, t) = \epsilon_0 \chi_{ijk}^{(2)} E_i(\mathbf{r}, t) E_j(\mathbf{r}, t). \tag{II.3}$$

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 ω_2. Defining

$$\mathbf{E}^{(-)}(\mathbf{r}, t) = \int \frac{d^2 k_\perp}{(2\pi)^2} \frac{d\omega}{2\pi} \mathbf{E}^{(-)}(\mathbf{k}_\perp, z, \omega) e^{-i\mathbf{k}_\perp \cdot \rho + i\omega t}, \tag{II.4}$$

[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

$$\partial_z \tilde{A}_1^{(+)}(\mathbf{k}_{1,\perp}, z, \omega) = \frac{2i\omega_1^2 d_{\text{eff}} A_3}{c^2 k_{1,z}(\omega_1)} \tilde{A}_2^{(-)}(\mathbf{k}_{3,\perp} - \mathbf{k}_{1,\perp}, z, \omega_3 - \omega) e^{i\Delta k_z z}, \tag{II.5}$$

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

$$\begin{aligned} A_1^{(+)}(\mathbf{k}_{1,\perp}, L, \omega) = &\left(\cosh(\lambda L) - \frac{i\Delta k_z}{2\lambda}\sinh(\lambda L)\right) A_1^{(+)}(\mathbf{k}_{1,\perp}, 0, \omega) \\ &+ \frac{\kappa_1}{\lambda}\sinh(\lambda L) A_2^{(-)}(\mathbf{k}_{2,\perp}, 0, \omega_3 - \omega), \end{aligned} \tag{II.6}$$

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

$$1 = \left| \left(\cosh(\lambda L) - \frac{i\Delta k_z}{2\lambda}\sinh(\lambda L)\right) \right|^2 - \left|\frac{\kappa_1}{\lambda}\right|^2 \sinh^2(\lambda L) R_C, \tag{II.7}$$

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]

$$R_C = \frac{n_1 \cos\theta_1 \omega_2}{n_2 \cos\theta_2 \omega_1}. \tag{II.8}$$

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

$$a_1(\mathbf{k}_{1,\perp}, L, \omega_1) = \mu(\mathbf{k}_{1,\perp}, \omega_1) a_1(\mathbf{k}_{1,\perp}, 0, \omega_1) + \nu(\mathbf{k}_{1,\perp}, \omega_1) a_2^\dagger(\mathbf{k}_{2,\perp}, 0, \omega_3 - \omega_1), \tag{II.9}$$

with

$$\mu(\mathbf{k}_{1,\perp}, \omega_1) = \cosh(\lambda L) - \frac{i\Delta k_z}{2\lambda}\sinh(\lambda L), \nu(\mathbf{k}_{1,\perp}, \omega_1) = i\sqrt{\frac{\kappa_1\kappa_2^*}{\lambda^2}}\sinh(\lambda L). \tag{II.10}$$

[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.

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 ω. 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].

A. 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 ω_1) is given by

$$n_1 = \sum_{\mathbf{k}_{1,\perp},\omega_1} \frac{\kappa_1\kappa_2^*}{\lambda^2}\sinh^2(\lambda L). \tag{II.11}$$

Since $\mathbf{k}_\perp, \omega$ are effectively continuous, we may write them as integrals as follows

$$n_1 = AT\int \frac{d^2k_{1,\perp}}{(2\pi)^2}\frac{d\omega}{(2\pi)}\frac{\kappa_1\kappa_2^*}{\lambda^2}\sinh^2(\lambda L), \tag{II.12}$$

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

$$\frac{dR}{d\omega} = A\int \frac{d^2k_{1,\perp}}{(2\pi)^3}\frac{\kappa_1\kappa_2^*}{\lambda^2}\sinh^2(\lambda L). \tag{II.13}$$

[6] You see why the delta function approach can be nice: no need for the large box!

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

$$\frac{1}{\hbar\omega_1}\frac{dI_1}{d\omega} = \int \frac{d^2k_{1,\perp}}{(2\pi)^3}\frac{4\omega_1^2\omega_2^2 d_{\text{eff}}^2|A_3|^2L^2}{k_{1,z}(\mathbf{k}_{1,\perp},\omega_1)k_{2,z}(\mathbf{k}_{2,\perp},\omega_2)c^4}\text{sinhc}^2\left(\sqrt{\frac{4\omega_1^2\omega_2^2 d_{\text{eff}}|A_3|^2L^2}{k_{1,z}k_{2,z}c^4} - \frac{(\Delta k_z)^2}{4}}L\right), \tag{II.14}$$

where I have defined 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

$$U = \exp\left[\zeta a^\dagger b^\dagger - \zeta^* ab\right], \tag{II.15}$$

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

$$|\psi\rangle = \bigotimes_{\mathbf{k}_\perp,\omega}\exp\left[\zeta_{\mathbf{k}_\perp,\omega}a^\dagger_{\mathbf{k}_\perp,\omega}b^\dagger_{-\mathbf{k}_\perp,(\omega_3-\omega)} - \zeta^*_{\mathbf{k}_\perp,\omega}a_{\mathbf{k}_\perp,\omega}b_{-\mathbf{k}_\perp,(\omega_3-\omega)}\right]|0\rangle. \tag{II.16}$$

If the nonlinearity is weak such that the expected number of photons is much less than 1, then we may approximate this as

$$|\psi\rangle \approx |0\rangle + \sum_{\mathbf{k}_\perp,\omega}\zeta_{\mathbf{k}_\perp,\omega}|1_{\mathbf{k}_\perp,\omega}1_{-\mathbf{k}_\perp,(\omega_3-\omega)}\rangle. \tag{II.17}$$

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].

1. 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 ϕ_1, ϕ_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

$$\text{sinc}^2\left(\frac{1}{2}(\Delta k_z)L\right), \tag{II.18}$$

where

$$\Delta k_z = k_{p,z} - k_{1,z} - k_{2,z}, \tag{II.19}$$

with

$$k_{i,z} = \sqrt{n_{s_i}^2(\omega_i, \theta)\frac{\omega_i^2}{c^2} - \mathbf{k}_\perp^2}, \tag{II.20}$$

where s_i is the polarization state of photon i. For a given c-axis orientation of a crystal (β-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

$$a^\dagger_{\mathbf{k}_\perp, o} a^\dagger_{-\mathbf{k}_\perp, o}|0\rangle = |\mathbf{k}o, -\mathbf{k}o\rangle. \tag{II.21}$$

[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.

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 polarization entanglement. In particular, we see that there is a contribution to the overall photon state of the form

$$|\psi\rangle \sim a^{\dagger}_{\mathbf{k}_{\perp},e} a^{\dagger}_{-\mathbf{k}_{\perp},o}|0\rangle + a^{\dagger}_{\mathbf{k}_{\perp},o} a^{\dagger}_{-\mathbf{k}_{\perp},e}|0\rangle = |\mathbf{k}e, -\mathbf{k}o\rangle + |\mathbf{k}o, -\mathbf{k}e\rangle, \tag{II.22}$$

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.

2. 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.

B. 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 ν 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 $\Delta k_z = 0$

$$a \to \mu a + \nu a^\dagger, \text{with } \mu = \cosh\lambda L, \nu = ie^{i\phi}\sinh\lambda L, \tag{II.23}$$

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

$$\frac{dA_1}{dz} = \frac{2i\omega_1 d_{\text{eff}}}{n_1 c} A_3 A_1^* e^{-i\Delta kz}; \; \frac{dA_3}{dz} = \frac{i\omega_3 d_{\text{eff}}}{n_2 c} A_1^2 e^{i\Delta kz}. \tag{II.24}$$

In the non-depleted pump approximation, we can treat A_3 as constant, which yields in the phase-matched case

$$A_1(L) = A_1(0)\cosh\lambda L + ie^{i\phi}A_1^*(0)\sinh\lambda L, \tag{II.25}$$

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

$$(\Delta X_\theta)^2 = \frac{1}{2}\left|\cosh(\lambda L)e^{-i\theta} - ie^{i\phi}\sinh(\lambda L)e^{i\theta}\right|^2, \tag{II.26}$$

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 λL significant without significant intensities, which lead to other complications.

III. 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 λ 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 ν 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 $a \to -r_1 a + it_1 b_{\text{in},1}$ 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 $a \to -r_2 a + it_2 b_{\text{in},2}$.

The combined effect of these transformations for the sth round trip can be written as

$$a_s \to a_{s+1} = r_1 r_2 \mu a_s + r_1 r_2 \nu a_s^\dagger - it_1 r_2 b_{\text{in},1,s} + it_2 b_{\text{in},2,s}. \tag{III.1}$$

To get to a differential equation, we would like to compute

$$\frac{a_{s+1} - a_s}{\tau} = \frac{1}{\tau}\left((r_1 r_2 \mu - 1)a_s + r_1 r_2 \nu a_s^\dagger - it_1 r_2 b_{\text{in},1,s} + it_2 b_{\text{in},2,s}\right), \tag{III.2}$$

where τ is the round-trip time. Now, we will approximate this as

$$\frac{1}{\tau}\left((r_1 r_2 - 1)a_s + i(\lambda L)a_s^\dagger - it_1 b_{\text{in},1,s} + it_2 b_{\text{in},2,s}\right), \tag{III.3}$$

where we have used $|r_{1,2}| \approx 1$ and neglected quadratic in λL 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

$$\frac{1}{\tau}\left(-\frac{1}{2}(|t_1|^2 + |t_2|^2)a_s + i(\lambda L)a_s^\dagger - it_1 b_{\text{in},1,s} + it_2 b_{\text{in},2,s}\right). \tag{III.4}$$

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

$$\frac{a_{s+1} - a_s}{\tau} \approx -\frac{1}{2}\gamma a_s + g a_s^\dagger - i\sqrt{\gamma_1}\frac{b_{\text{in},1,s}}{\sqrt{\tau}} + i\sqrt{\gamma_2}\frac{b_{\text{in},2,s}}{\sqrt{\tau}}. \tag{III.5}$$

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

$$\left[\frac{b_{\text{in},i,s}}{\sqrt{\tau}}, \frac{b^\dagger_{\text{in},j,s'}}{\sqrt{\tau}}\right] = \delta_{ij}\frac{\delta_{s,s'}}{\tau}. \tag{III.6}$$

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

$$\frac{b_{\text{in},i}(t)}{\sqrt{\tau}} \to b_{\text{in},i}(t). \tag{III.7}$$

We may now express the commutator in the $\tau \to 0$ limit as

$$[b_{\text{in},i}(t), b^\dagger_{\text{in},j}(t)] = \delta_{ij}\delta(t - t'). \tag{III.8}$$

From now on, if we write b_{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

$$\dot{a} = -\frac{1}{2}\gamma a + g a^\dagger + F. \tag{III.9}$$

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

$$\dot{a}^\dagger = -\frac{1}{2}\gamma a^\dagger + g^* a + F^\dagger. \tag{III.10}$$

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

$$b_{\mathrm{out},i}(t) = -r b_{\mathrm{in},i}(t) + \frac{it_i}{\sqrt{\tau}} a(t) \approx -b_{\mathrm{in},i}(t) + i\sqrt{\gamma_i}\, a(t). \tag{III.11}$$

This equation of motion for the intracavity field operator can be solved by standard (Fourier) techniques. Fourier transforming, we have

$$\begin{aligned} -i\omega a(\omega) &= -\frac{1}{2}\gamma a(\omega) + g a(-\omega)^\dagger + F(\omega) \\ -i\omega a(-\omega)^\dagger &= -\frac{1}{2}\gamma a(-\omega)^\dagger + g^* a(\omega) + F(-\omega)^\dagger, \end{aligned} \tag{III.12}$$

where I have used $\int dt\; e^{i\omega t} A(\omega) = (A(-\omega))^\dagger$. This matrix equation is solved by

$$\begin{pmatrix} a(\omega) \\ a(-\omega)^\dagger \end{pmatrix} = \left(\left(\frac{1}{2}\gamma - i\omega\right)^2 - |g|^2 \right)^{-1} \begin{pmatrix} \frac{1}{2}\gamma - i\omega & g \\ g^* & \frac{1}{2}\gamma - i\omega \end{pmatrix} \begin{pmatrix} F(\omega) \\ F(-\omega)^\dagger . \end{pmatrix} \tag{III.13}$$

A. 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

[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.

pump phase such that g is real. A different phase of ϕ 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

$$[b_{\text{in}}(\omega), b_{\text{in}}^{\dagger}(\omega')] = 2\pi\delta(\omega - \omega'). \tag{III.14}$$

The output field operator in frequency space is given by

$$\chi^{bb}(\omega) = i\sqrt{\gamma_1}a(\omega) - b_{\text{in}}(\omega) \equiv \chi^{bb}(\omega)b_{\text{in}}(\omega) + \chi^{bb^{\dagger}}(\omega)b_{\text{in}}(-\omega)^{\dagger}, \tag{III.15}$$

where

$$\chi^{bb}(\omega) = i\sqrt{\gamma_1}\left(\frac{-i\sqrt{\gamma_1}\left(\frac{1}{2}\gamma_1 - i\omega\right)}{\left(\frac{1}{2}\gamma_1 - i\omega\right)^2 - g^2}\right) - 1 = \frac{\frac{\gamma_1^2}{4} + \omega^2 + g^2}{\left(\frac{1}{2}\gamma_1 - i\omega\right)^2 - g^2}. \tag{III.16}$$

and

$$\chi^{bb^{\dagger}}(\omega) = -\frac{\gamma_1 g}{\left(\frac{1}{2}\gamma_1 - i\omega\right)^2 - g^2}. \tag{III.17}$$

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_θ^{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]:

$$S_\theta^{XX} = \langle X(\omega)^{\dagger} X(\omega)\rangle. \tag{III.18}$$

To evaluate this, let's write

$$X(\omega) = (\chi^{bb}(\omega)b_{\text{in}}(\omega) + \chi^{bb^{\dagger}}(\omega)b_{\text{in}}(-\omega)^{\dagger})e^{-i\theta} + (\chi^{bb*}(-\omega)b_{\text{in}}(-\omega)^{\dagger} + \chi^{bb^{\dagger}*}(-\omega)b_{\text{in}}(\omega))e^{i\theta} \tag{III.19}$$

[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.

and regroup terms as

$$X(\omega) = (\chi^{bb}(\omega)e^{-i\theta} + \chi^{bb^\dagger *}(-\omega)e^{i\theta})b_{\text{in}}(\omega) + (\chi^{bb^\dagger}(\omega)e^{-i\theta} + \chi^{bb*}(-\omega)e^{i\theta})b_{\text{in}}(-\omega)^\dagger. \tag{III.20}$$

Assuming vacuum statistics for the input fields, it follows that the squeezing spectrum is given by

$$S_\theta^{XX}(\omega) = \left|\chi^{bb^\dagger}(\omega)e^{-i\theta} + \chi^{bb*}(-\omega)e^{i\theta}\right|^2. \tag{III.21}$$

For our particular system, we have

$$S_\theta^{XX}(\omega) = \left|\frac{-\gamma_1 g e^{-i\theta} + (\frac{\gamma_1^2}{4} + \omega^2 + g^2)e^{i\theta}}{\left(\frac{1}{2}\gamma_1 - i\omega\right)^2 - g^2}\right|^2 \tag{III.22}$$

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

$$S_0^{XX} = \frac{\left(\frac{\gamma_1}{2} - g\right)^2 + \omega^2}{\left(\frac{\gamma_1}{2} + g\right)^2 + \omega^2}, \tag{III.23}$$

while in the orthogonal quadrature, the spectrum is given as:

$$S_{\pi/2}^{XX} = \frac{\left(\frac{\gamma_1}{2} + g\right)^2 + \omega^2}{\left(\frac{\gamma_1}{2} - g\right)^2 + \omega^2}. \tag{III.24}$$

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 ω 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

$$-\frac{1}{2}\gamma_1 I + \begin{pmatrix} 0 & g \\ g^* & 0 \end{pmatrix} \tag{III.25}$$

is non-invertible, which requires a vanishing of the determinant:

$$\frac{\gamma_1^2}{4} - |g|^2 \implies |g| = \frac{\gamma_1}{2}. \tag{III.26}$$

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 $g > \gamma_1/2$.

B. 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:

$$(\Delta X)^2 = \frac{1}{2}\langle (a + a^\dagger)^2 \rangle, (\Delta P)^2 = -\frac{1}{2}\langle (a - a^\dagger)^2 \rangle, \tag{III.27}$$

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

$$X(t) = \int \frac{d\omega}{2\pi} e^{-i\omega t} X(\omega), \tag{III.28}$$

[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.

where $X(\omega) = \frac{1}{\sqrt{2}}\left(a(\omega) + a(-\omega)^\dagger\right)$. Therefore

$$(\Delta X)^2 = \int \frac{d\omega}{2\pi}\frac{d\omega'}{2\pi} e^{-i(\omega+\omega')t}\langle X(\omega)X(\omega')\rangle. \tag{III.29}$$

Let us now consider the quantity $\langle X(\omega)X(\omega')\rangle$ which is given by:

$$\left\langle \left(\chi^{XF}(\omega)F(\omega) + \chi^{XF^\dagger}(\omega)F(-\omega)^\dagger\right)\left(\chi^{XF}(\omega')F(\omega') + \chi^{XF^\dagger}(\omega')F(-\omega')^\dagger\right)\right\rangle \tag{III.30}$$

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_{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 $[b_{\mathrm{in}}(\omega), b^\dagger_{\mathrm{in}}(\omega')] = 2\pi\delta(\omega-\omega')$. We see then that

$$(\Delta X)^2 = \gamma_1 \int \frac{d\omega}{2\pi}\chi^{XF}(\omega)\chi^{XF^\dagger}(-\omega). \tag{III.31}$$

Using

$$\chi^{XF}(\omega) = \frac{1}{\sqrt{2}}\frac{\frac{1}{2}\gamma_1 - i\omega + g}{\left(\frac{1}{2}\gamma_1 - i\omega\right)^2 - g^2} = \frac{1}{\sqrt{2}}\frac{1}{\frac{1}{2}\gamma_1 - i\omega - g} \tag{III.32}$$

and

$$\chi^{XF^\dagger}(-\omega) = \frac{1}{\sqrt{2}}\frac{\frac{1}{2}\gamma_1 + i\omega + g}{\left(\frac{1}{2}\gamma_1 + i\omega\right)^2 - g^2} = \frac{1}{\sqrt{2}}\frac{1}{\frac{1}{2}\gamma_1 + i\omega - g}, \tag{III.33}$$

we have

$$(\Delta X)^2 = \frac{1}{2}\int \frac{d\omega}{2\pi}\frac{1}{\left(\frac{1}{2}\gamma_1 - g\right)^2 + \omega^2} = \frac{1}{4}\frac{\gamma_1}{\frac{1}{2}\gamma_1 - g} = \frac{1}{2}\frac{1}{1 - \frac{2g}{\gamma_1}}. \tag{III.34}$$

The P variance follows immediately from its definition as

$$(\Delta P)^2 = \frac{1}{2}\frac{1}{1 + \frac{2g}{\gamma_1}}. \tag{III.35}$$

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.

IV. 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 ω_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

$$E(z,t) = A^{(+)}(z,t)e^{ik(\omega_1)z-i\omega_1 t} + A^{(-)}(z,t)e^{-ik(\omega_1)z+i\omega_1 t}. \tag{IV.1}$$

A very similar derivation to the classical case then yields the Heisenberg equation

$$\partial_z \tilde{A}_1^{(+)}(z,t) = \sum_{m=1}^{\infty} \frac{i^{m+1}\beta_m}{m!}\partial_t^m \tilde{A}_1^{(+)}(z,t) + \frac{i\omega_1^2}{2\epsilon_0 c^2 k(\omega_1)}\tilde{P}_1^{(+)}(z,t). \tag{IV.2}$$

The relevant polarization at third-order is given by

$$\tilde{P}_1^{(+)}(z,t) = \epsilon_0\chi^{(3)}E(z,t)^3. \tag{IV.3}$$

Since we are only interested in the part of the polarization near frequency ω, we may take only the terms in E^3 that multiply $e^{ik(\omega_1)z-i\omega_1 t}$. Those terms are

$$\left(\tilde{A}_1^{(+)}(z,t)\tilde{A}_1^{(+)}(z,t)\tilde{A}_1^{(-)}(z,t) + \tilde{A}_1^{(+)}(z,t)\tilde{A}_1^{(-)}(z,t)\tilde{A}_1^{(+)}(z,t) + \tilde{A}_1^{(-)}(z,t)\tilde{A}_1^{(+)}(z,t)\tilde{A}_1^{(+)}(z,t)\right). \tag{IV.4}$$

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:

$$[A^{(+)}(z,t),A^{(-)}(z',t')] = [E^{(+)}(z,t),E^{(-)}(z',t')]e^{-ik(\omega_1)(z-z')+i\omega_1(t-t')}. \tag{IV.5}$$

The electric field operator, in our quasi-1D, scalar case can be written as an expansion in plane waves as

$$E^{(+)}(z,t) = i\int_{\Delta k}\frac{dk}{2\pi}\mathcal{E}(k)a(k,t)e^{ikz-i\omega_k t}, \tag{IV.6}$$

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

$$\mathcal{E}(k) = \sqrt{\frac{\hbar\omega(k)}{2\epsilon_0 n^2 A}}, \tag{IV.7}$$

with n an effective mode index and A an effective mode area. We restrict the mode expansion to a wavenumber bandwidth Δk since this simplified description of the field may not be valid everywhere. Therefore, the commutator can be written as

$$C^E(z-z',t-t') = [E^{(+)}(z,t),E^{(-)}(z',t')] = \int_{\Delta k}\frac{dk}{2\pi}|\mathcal{E}(k)|^2 e^{ik(z-z')-i\omega_k(t-t')}. \tag{IV.8}$$

The equal time commutator is given by

$$C^E(z-z',0) = \int_{\Delta k}\frac{dk}{2\pi}|\mathcal{E}(k)|^2 e^{ik(z-z')}. \tag{IV.9}$$

Let us suppose that the bandwidth Δk 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 $\Delta k/2$:

$$C^E(z-z',0) \approx |\mathcal{E}(k_0)|^2\frac{\Delta k}{2\pi}\text{sinc}\left(\frac{\Delta k}{2}(z-z')\right). \tag{IV.10}$$

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

$$3\tilde{A}_1^{(-)}(z,t)\tilde{A}_1^{(+)}(z,t)\tilde{A}_1^{(+)}(z,t) + 3A^{(+)}(z,t)C^E(0,0). \tag{IV.11}$$

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

$$\partial_z \tilde{A}_1^{(+)}(z,t) = \sum_{m=1}^{\infty} \frac{i^{m+1}\beta_m}{m!} \partial_t^m \tilde{A}_1^{(+)}(z,t) + \frac{3i\omega_1\chi^{(3)}}{2cn_1(\omega)} \tilde{A}_1^{(-)}(z,t)\tilde{A}_1^{(+)}(z,t)^2. \quad \text{(IV.12)}$$

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

$$A^{(+)}(z,t) = \sqrt{\frac{\hbar\omega_1}{2\epsilon_0 n^2 A}}\psi(z,t); A^{(-)}(z,t) = \sqrt{\frac{\hbar\omega_1}{2\epsilon_0 n^2 A}}\psi^\dagger(z,t) \quad \text{(IV.13)}$$

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

$$\partial_z \psi(z,t) = \sum_{m=1}^{\infty} \frac{i^{m+1}\beta_m}{m!} \partial_t^m \psi(z,t) + i\kappa\psi^\dagger(z,t)\psi^2(z,t), \quad \text{(IV.14)}$$

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

$$\partial_{z'} \psi(z',t') = \sum_{m=2}^{\infty} \frac{i^{m+1}\beta_m}{m!} \partial_t^m \psi(z',t') + i\kappa\psi^\dagger(z',t')\psi^2(z',t'). \quad \text{(IV.15)}$$

The commutator at equal z can *approximately* be written as

$$C^\psi(0, t_1' - t_2') \approx \frac{\Delta k}{2\pi} \text{sinc}\left(\frac{v_g \Delta k}{2}(t_1' - t_2')\right). \quad \text{(IV.16)}$$

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 ψ to be a flux of photons (photons per unit time). This is done be rescaling ψ 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:

$$\partial_{z'}\psi(z',t') = \sum_{m=2}^{\infty} \frac{i^{m+1}\beta_m}{m!}\partial_t^m \psi(z',t') + i\kappa\psi^\dagger(z',t')\psi^2(z',t'). \tag{IV.17}$$

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 κ parameter is related to the γ 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 κ is the constant for photon-number-per-unit-time-normalized fields while γ is the constant for energy-per-unit-time normalized fields.

A. 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

$$\partial_z \psi(z,t') = i\kappa\psi^\dagger(z,t')\psi^2(z,t'), \tag{IV.18}$$

where I have taken $z' \to z$ since their meaning is the same. This equation is exactly solvable. It uses the fact that

$$\partial_z \psi^\dagger(z,t')\psi(z,t') = \psi^\dagger(z,t')(i\kappa\psi^\dagger(z,t')\psi^2(z,t')) - (i\kappa\psi^{\dagger 2}(z,t')\psi(z,t'))\psi(z,t') = 0. \tag{IV.19}$$

Therefore, we may write our Heisenberg equation as

$$\partial_z \psi(z,t') = i\kappa(\psi^\dagger(0,t')\psi(0,t'))\psi(z,t'), \tag{IV.20}$$

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

$$\psi(z,t') = e^{i\kappa z\psi^\dagger(0,t')\psi(0,t')}\psi(0,t'). \tag{IV.21}$$

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

$$a \equiv \sqrt{\Delta t}\psi(t), \tag{IV.22}$$

where Δt is some temporal bin width. This allows us to associate the self-phase modulation with a transformation

$$a \to e^{i\theta a^\dagger a} a, \tag{IV.23}$$

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 Δt. 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 Δt 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 δa represents the quantum fluctuations of the operator and α 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 δa. 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 2π. But if we consider

$$\theta a^\dagger a = \theta\left(|\alpha|^2 + (\alpha^*\delta a + \alpha\delta a^\dagger) + \delta a^\dagger \delta a\right), \tag{IV.24}$$

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 2π 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 δa, leaving us with

$$a \to e^{i\theta a^\dagger a} a \approx e^{i\theta|\alpha|^2}\left(1 + i\theta(\alpha^*\delta a + \alpha\delta a^\dagger)\right)(\alpha + \delta a). \tag{IV.25}$$

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

$$a \to e^{i\theta|\alpha|^2}\left(\alpha + \left(1 + i\theta|\alpha|^2\right)\delta a + i\theta\alpha^2\delta a^\dagger\right). \tag{IV.26}$$

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

$$\alpha \to e^{i\theta|\alpha|^2}\alpha, \tag{IV.27}$$

which is the usual SPM transformation. The fluctuations transform as

$$\delta a \to \mu\delta a + \nu\delta a^\dagger, \text{where } \mu = \left(1 + i\theta|\alpha|^2\right)e^{i\theta|\alpha|^2}, \nu = i\theta\alpha^2 e^{i\theta|\alpha|^2} \tag{IV.28}$$

We immediately see that $|\mu|^2 - |\nu|^2 = 1$ as we expect for the Bogoliubov transformation. The ζ parameter for the transformation, or equivalently $|r|$ and ϕ 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

$$b' = D^\dagger(\alpha)S^\dagger(\zeta)bS(\zeta)D(\alpha) = D^\dagger(\alpha)(\mu b+\nu b^\dagger)D(\alpha) = \mu(b+\alpha)+\nu(b^\dagger+\alpha^*), \quad \text{(IV.29)}$$

where μ, ν are determined from ζ 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

$$\delta b' = \mu(b+\alpha) + \nu(b^\dagger + \alpha^*) - (\mu\alpha + \nu\alpha^*) = \mu b + \nu b^\dagger \quad \text{(IV.30)}$$

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 δb coincide. Therefore we have

$$\delta b' = \mu\delta b + \nu\delta b^\dagger, \quad \text{(IV.31)}$$

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.

B. 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)

$$|\psi'\rangle = e^{-i\theta N} e^{i\theta N^2} |\psi(0)\rangle \tag{IV.32}$$

where $\theta = -Kt/2$. Suppose that we have a coherent state as our initial state. Then, employing its photon number representation, we see that

$$|\psi'\rangle = e^{-\frac{1}{2}|\alpha|^2} \sum_{n=0}^{\infty} e^{i\theta n^2} \frac{(\alpha e^{-i\theta})^n}{\sqrt{n!}} |n\rangle. \tag{IV.33}$$

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

$$e^{in^2\theta} = \frac{1}{\sqrt{2}} e^{i\frac{\pi}{4}} + (-1)^n e^{-i\frac{\pi}{4}}. \tag{IV.34}$$

Therefore it immediately follows that the resulting state is

$$|\psi'\rangle = \frac{1}{\sqrt{2}} \left(e^{i\frac{\pi}{4}} |\alpha e^{-i\theta}\rangle + e^{-i\frac{\pi}{4}} |-\alpha e^{-i\theta}\rangle \right), \tag{IV.35}$$

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.

V. 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:

$$\begin{aligned}\dot{\mathbf{a}} &= F(\mathbf{a}, \mathbf{a}^\dagger) \\ \dot{\mathbf{a}}^\dagger &= F^\dagger(\mathbf{a}, \mathbf{a}^\dagger),\end{aligned} \tag{V.1}$$

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

$$\mathbf{a} = \boldsymbol{\alpha} + \delta\mathbf{a}, \tag{V.2}$$

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

$$\dot{\boldsymbol{\alpha}} = F(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*), \tag{V.3}$$

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

$$\delta \dot{a}_i = \sum_j \frac{\partial F_i}{\partial \alpha_j} \delta a_j + \sum_j \frac{\partial F_i}{\partial \alpha_j^*} \delta a_j^\dagger \tag{V.4}$$

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:

$$\delta a_i(t) = \sum_j \mu_{ij} \delta a_j(0) + \nu_{ij} \delta a_j^\dagger(0). \tag{V.5}$$

In what follows, we will make a new connection, showing that the μ, ν 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. V.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

$$.\delta \dot{\alpha}_i = \sum_j \frac{\partial F_i}{\partial \alpha_j} \delta \alpha_j + \sum_j \frac{\partial F_i}{\partial \alpha_j^*} \delta \alpha_j^* \tag{V.6}$$

This equation is in correspondence with Eq. V.4, and its solution is the same, as Eq. V.4 is a linear equation, and thus is solved identically to a classical equation. In particular, we may write:

$$\delta \alpha_i(t) = \sum_j \mu_{ij} \delta \alpha_j(0) + \nu_{ij} \delta \alpha_j^\dagger(0), \tag{V.7}$$

where μ, ν are identical to the quantum-mechanical case. But, by construction, we may also write

$$\delta \alpha_i(t) = \sum_j \frac{\partial \alpha_i(t)}{\partial \alpha_j(0)} \delta \alpha_j(0) + \frac{\partial \alpha_i(t)}{\partial \alpha_j^*(0)} \delta \alpha_j^*(0), \tag{V.8}$$

allowing us to identify

$$\mu_{ij} = \frac{\partial \alpha_i(t)}{\partial \alpha_j(0)}, \nu_{ij} = \frac{\partial \alpha_i(t)}{\partial \alpha_j^*(0)}, \tag{V.9}$$

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 δX which is linear in fluctuation operators δa_i and $\delta a_i^\dagger$:

$$\delta X = \sum_i c_i \delta a_i + d_i \delta a_i^\dagger. \tag{V.10}$$

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

$$\begin{aligned}
\delta X(t) &= \sum_{ij} \left[c_i \frac{\partial \alpha_i(t)}{\partial \alpha_j(0)} + d_i \frac{\partial \alpha_i^*(t)}{\partial \alpha_j(0)} \right] \delta a_j(0) + \left[c_i \frac{\partial \alpha_i(t)}{\partial \alpha_j^*(0)} + d_i \frac{\partial \alpha_i^*(t)}{\partial \alpha_j^*(0)} \right] \delta a_j^\dagger(0) \\
&= \sum_j \frac{\partial X(t)}{\partial \alpha_j(0)} \delta a_j(0) + \frac{\partial X(t)}{\partial \alpha_j^*(0)} \delta a_j^\dagger(0) \\
&\equiv \left[\delta \boldsymbol{a} \cdot \frac{\partial}{\partial \boldsymbol{\alpha}} + \delta \boldsymbol{a}^\dagger \cdot \frac{\partial}{\partial \boldsymbol{\alpha}^*} \right] X(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*)
\end{aligned} \tag{V.11}$$

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:

$$\begin{aligned}
(\Delta X)^2 &= v^T C v, \text{where} \\
v &= \begin{pmatrix} \frac{\partial X}{\partial \boldsymbol{\alpha}} & \frac{\partial X}{\partial \boldsymbol{\alpha}^*} \end{pmatrix}^T \\
C &= \begin{pmatrix} \langle \delta \mathbf{a} \delta \mathbf{a} \rangle & \langle \delta \mathbf{a} \delta \mathbf{a}^\dagger \rangle \\ \langle \delta \mathbf{a}^\dagger \delta \mathbf{a} \rangle & \langle \delta \mathbf{a}^\dagger \delta \mathbf{a}^\dagger \rangle \end{pmatrix}
\end{aligned} \tag{V.12}$$

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.

A. 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.

1. 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

$$\begin{aligned} \dot{\alpha} &= \Omega\alpha^* \\ \dot{\alpha}^* &= \Omega^*\alpha. \end{aligned} \tag{V.13}$$

The solution is

$$\begin{pmatrix} \alpha(t) \\ \alpha^*(t) \end{pmatrix} = \exp\left[\begin{pmatrix} 0 & \Omega t \\ \Omega^* t & 0 \end{pmatrix}\right]\begin{pmatrix} \alpha(0) \\ \alpha^*(0) \end{pmatrix} = \begin{pmatrix} \cosh\left(|\Omega|t\right)\alpha(0) + \sinh(|\Omega|t)e^{i\phi}\alpha^*(0) \\ \sinh(|\Omega|t)e^{-i\phi}\alpha(0) + \cosh\left(|\Omega|t\right)\alpha^*(0) \end{pmatrix}, \tag{V.14}$$

where we have defined $\Omega = |\Omega|e^{i\phi}$. According to Eq. (S5), we can write the fluctuation operators as

$$\delta a(t) = \mu\delta a(0) + \nu\delta a^\dagger(0), \tag{V.15}$$

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:

$$(\Delta X_\varphi)^2 = \left|\frac{\partial X_\varphi(t)}{\partial a(0)}\right|^2 = \left|\cosh\left(|\Omega|t\right)e^{i\varphi} + \sinh(|\Omega|t)e^{-i\phi}e^{-i\varphi}\right|^2, \tag{V.16}$$

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

$$\alpha = \beta + \mu\alpha(0) + \nu\alpha^*(0). \tag{V.17}$$

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.

2. 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

$$\partial_z\alpha(z,t) = i\gamma\alpha^*(z,t)\alpha^2(z,t), \tag{V.18}$$

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

[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).

zero). Dimensions of α 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 γ is related to the nonlinear propagation phase. In particular, the pulse envelope, after a propagation distance L in the fiber, is given by:

$$\alpha(L,t) = e^{i\theta\alpha^*(0,t)\alpha(0,t)}\alpha(0,t), \tag{V.19}$$

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

$$\delta a(L,t) = \int ds \, \left[\frac{\partial\alpha(L,t)}{\partial\alpha(0,s)}\delta a(0,s) + \frac{\partial\alpha(L,t)}{\partial\alpha^*(0,s)}\delta a^\dagger(0,s)\right], \tag{V.20}$$

where

$$\frac{\partial\alpha(L,t)}{\partial\alpha(0,s)} = \delta(t-s)\left(1 + i\theta|\alpha(0,t)|^2\right)e^{i\theta|\alpha|^2(0,t)} \equiv \mu(s)\delta(t-s), \tag{V.21}$$

and

$$\frac{\partial\alpha(L,t)}{\partial\alpha^*(0,s)} = \delta(t-s)\left(i\theta\alpha(0,t)^2\right)e^{i\theta|\alpha|^2(0,t)} \equiv \nu(s)\delta(t-s), \tag{V.22}$$

where we have defined μ and ν for ease of notation. This relation is in exact agreement with the standard treatment based on an exact solution of the Heisenberg equations.