

Light-matter interactions with photonic quasiparticles

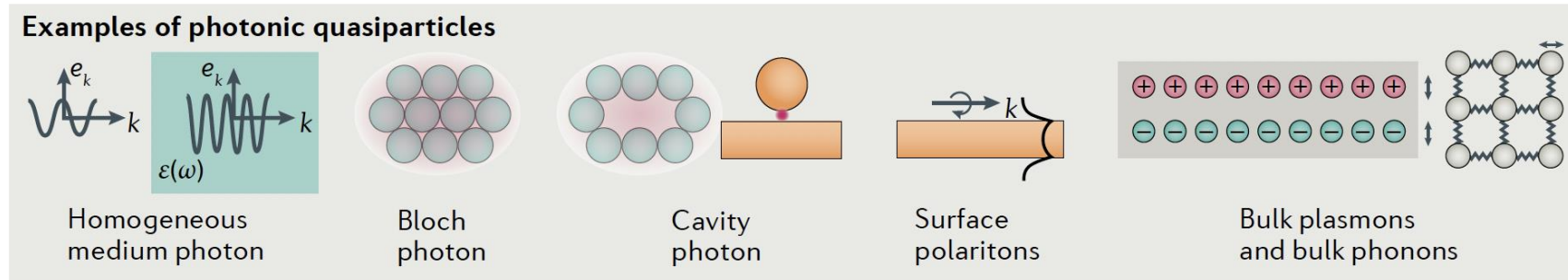
Nick Rivera

Massachusetts Institute of Technology

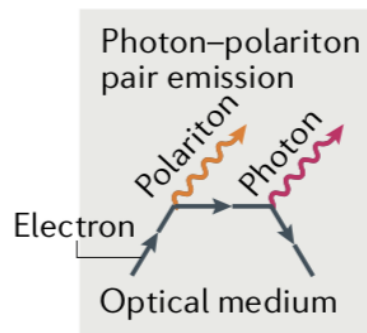
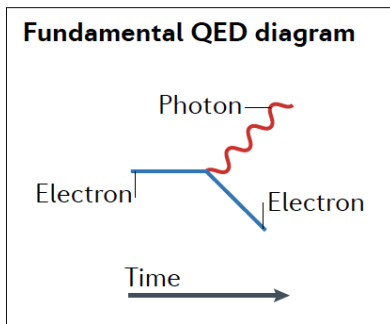
<https://nrivera.scripts.mit.edu/nhr>

Outline

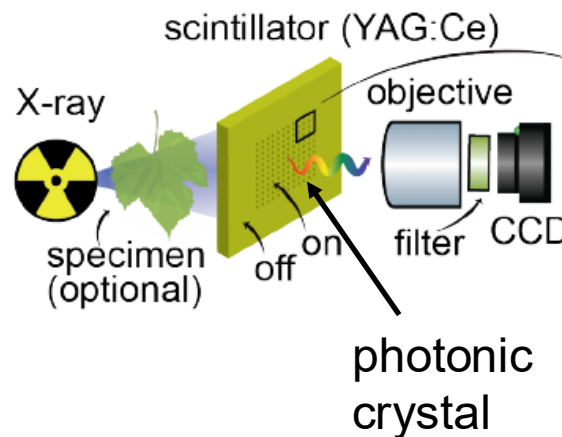
Photonic quasiparticles



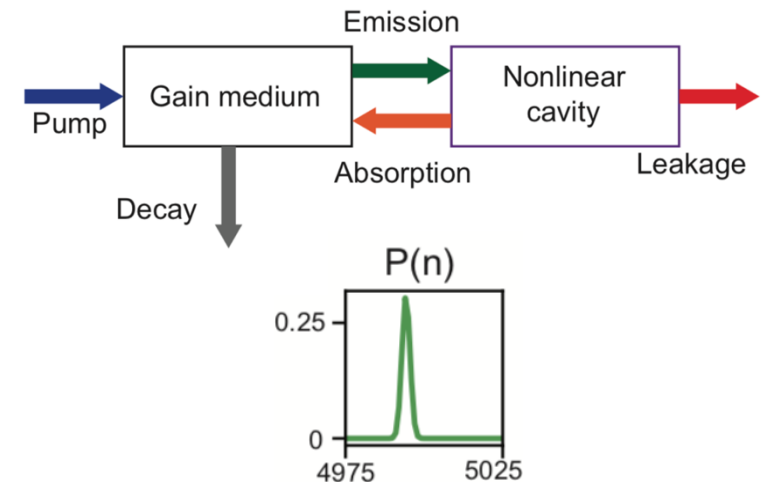
New light-matter interactions based on photonic quasiparticles



New particle detectors based on photonic quasiparticle interactions



Using photonic quasiparticle nonlinearities to generate macroscopic Fock states

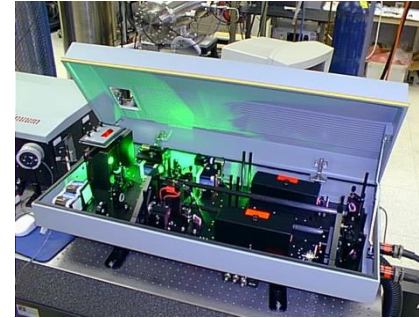


Light-matter interactions

Working definition: electronic transitions (bound and free electrons) leading to absorption, emission, or scattering of the photon.



Lasers



LEDs



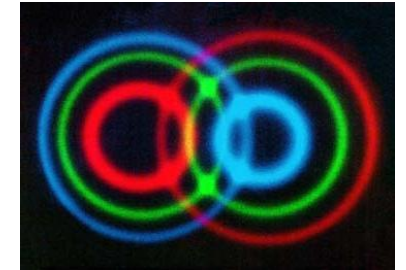
Solar cells



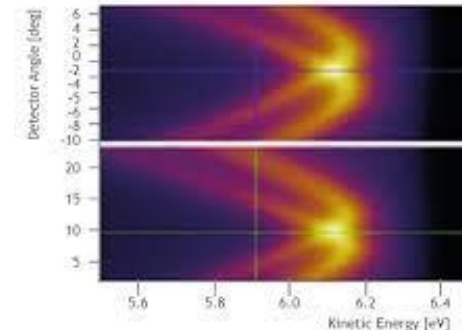
Detectors



Quantum light



Spectroscopy



Particle detection



Limitations of light-matter interactions

The interactions are *very weak*, are largely described at leading order.

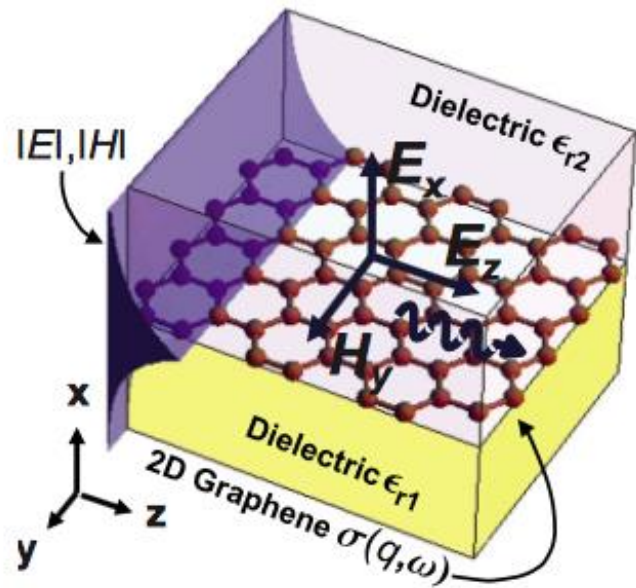
- Coupling strength: α
- Ratio of atomic size to optical wavelength: $ka = 2\pi a/\lambda$
- Scale of applied field to internal field: E_{ext}/E_0

All of these together, strongly limit the diversity of allowed interactions.

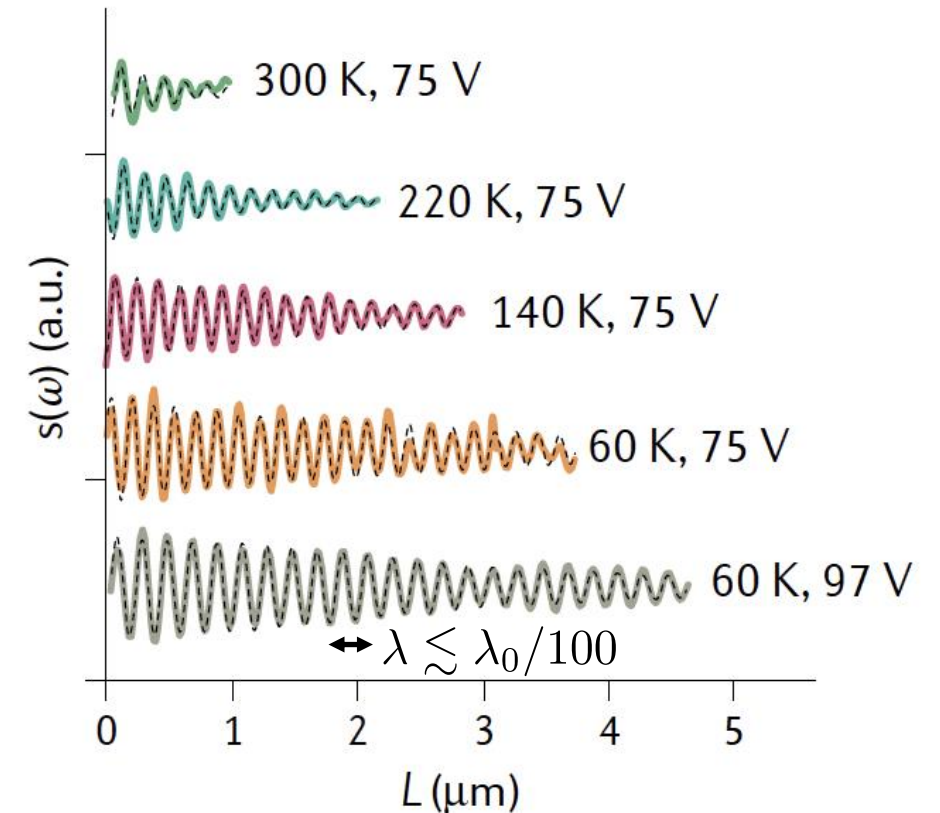
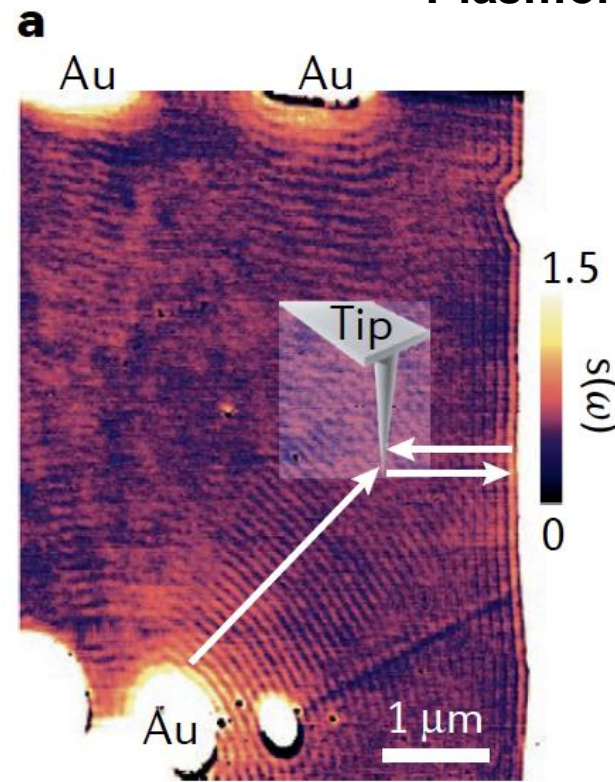
- Neglect “multipole” decays [dipole approx.]
- Neglect “multiphoton” decays [first-order QED approx.]
- Neglect nonlinear phenomena [first-order external field approx.]

Collective excitations in solids challenge these limitations

Plasmons in graphene

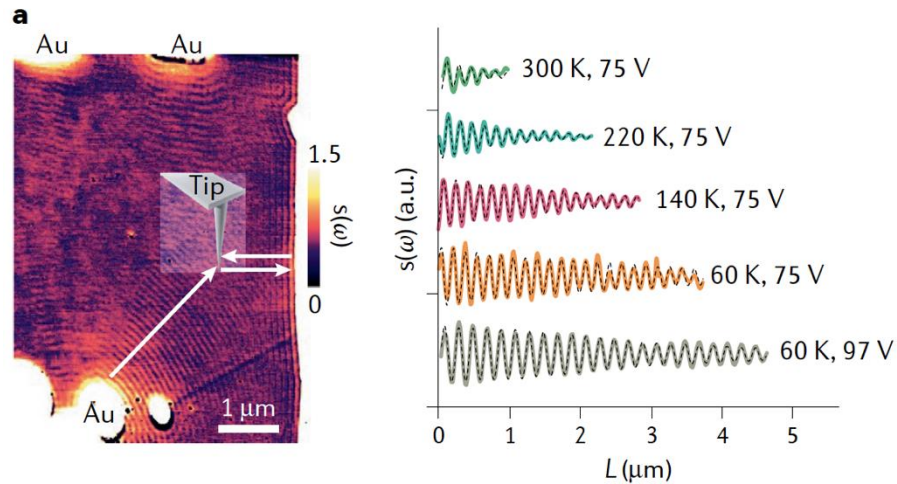


Jablan *et al.* *PRB* (2009).

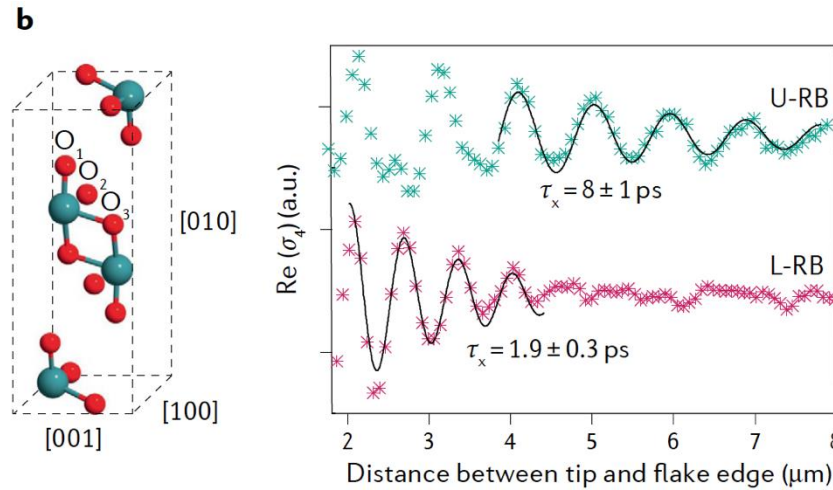


Ni *et al.* *Nature* (2018).

Collective excitations: like photons but with very different linear properties

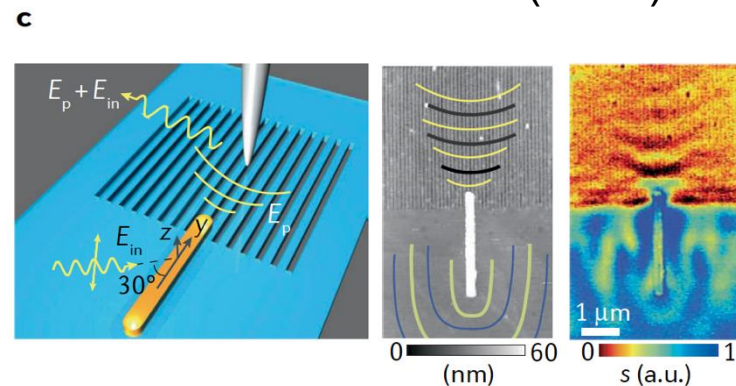


Ni et al. Nature (2018).

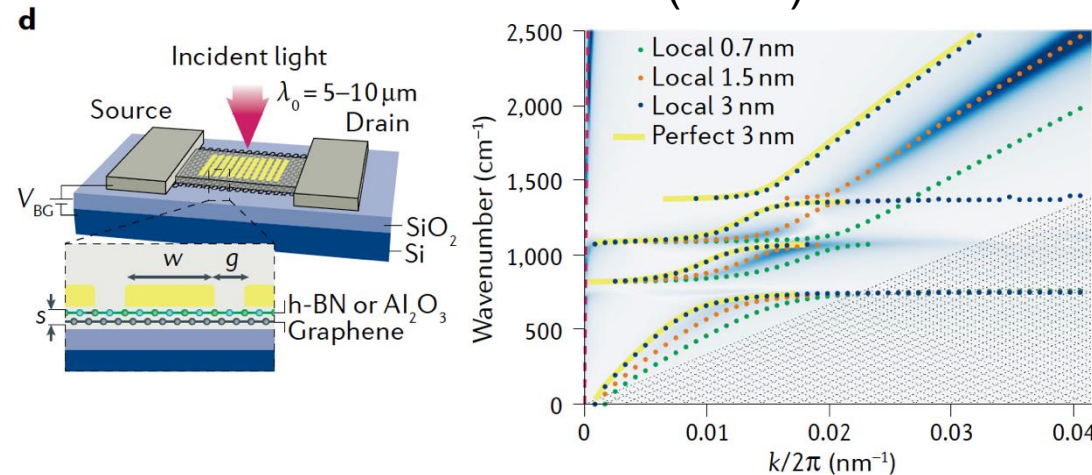


Ma et al. Nature (2018).

- *Wavelength* – $>100\times$ smaller
- *Polarization* – often circular
- *DOS* – very high
- *Mode volume* – very small



Li et al. Science (2018).



Iranzo et al. Science (2018).

Photonic quasiparticles (PQPs) as quantized solutions to Maxwell equations

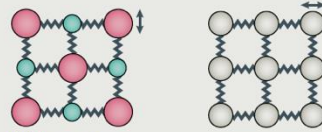
Microscopic origin of electromagnetic response



Free charges
(such as in metals,
Cooper pairs)



Bound electrons
(such as excitons,
atomic transitions)

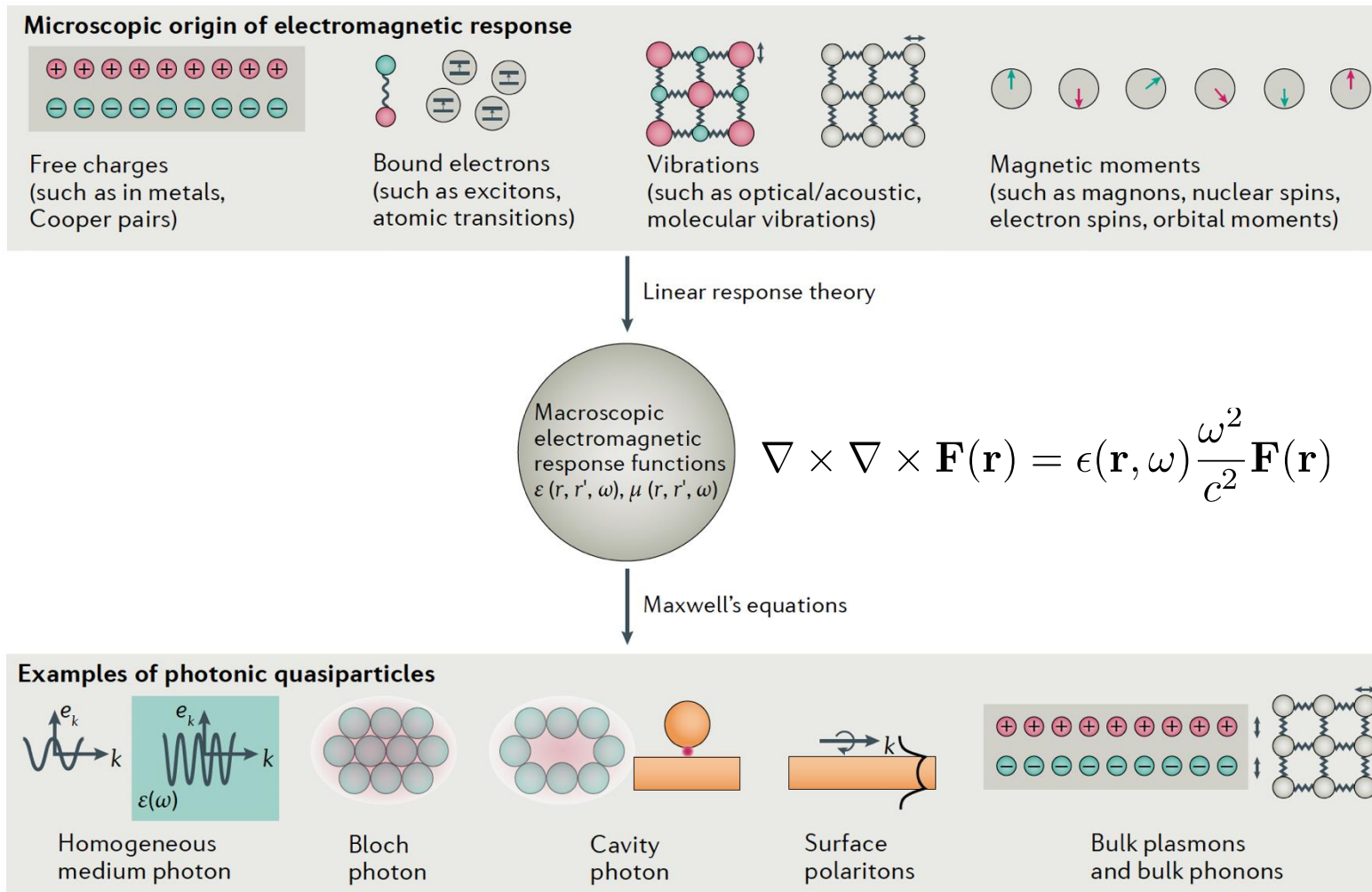


Vibrations
(such as optical/acoustic,
molecular vibrations)



Magnetic moments
(such as magnons, nuclear spins,
electron spins, orbital moments)

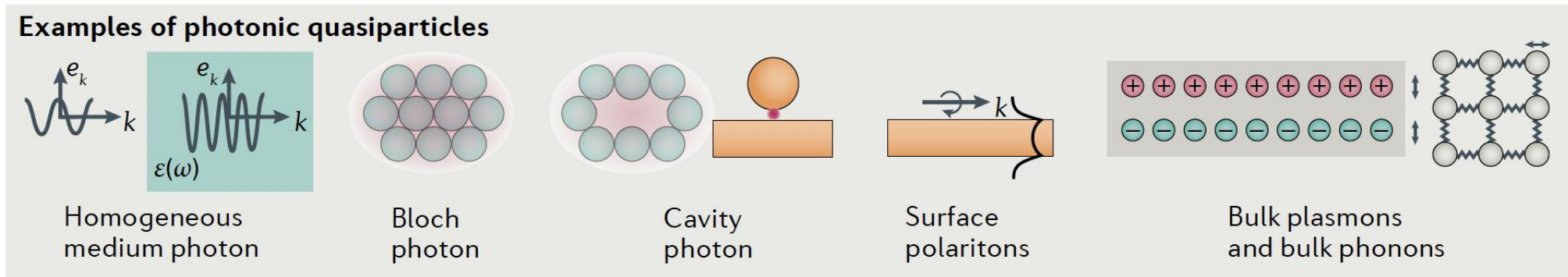
Photonic quasiparticles (PQPs) as quantized solutions to Maxwell equations



Quantized PQP field:

$$\mathbf{E}(\mathbf{r}) = i \sum_n \sqrt{\frac{\hbar \omega_n}{2\epsilon_0}} (\mathbf{F}_n(\mathbf{r}) a_n - \text{h.c.})$$

Light-matter interactions with photonic quasiparticles

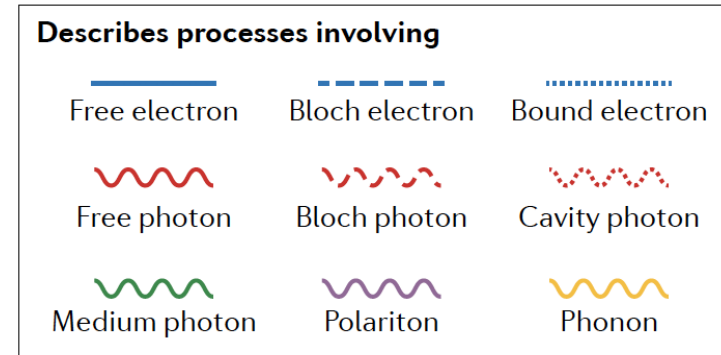
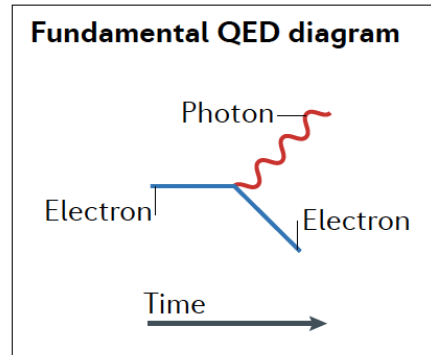


What happens when an excited electron interacts electromagnetically with photonic quasiparticles?

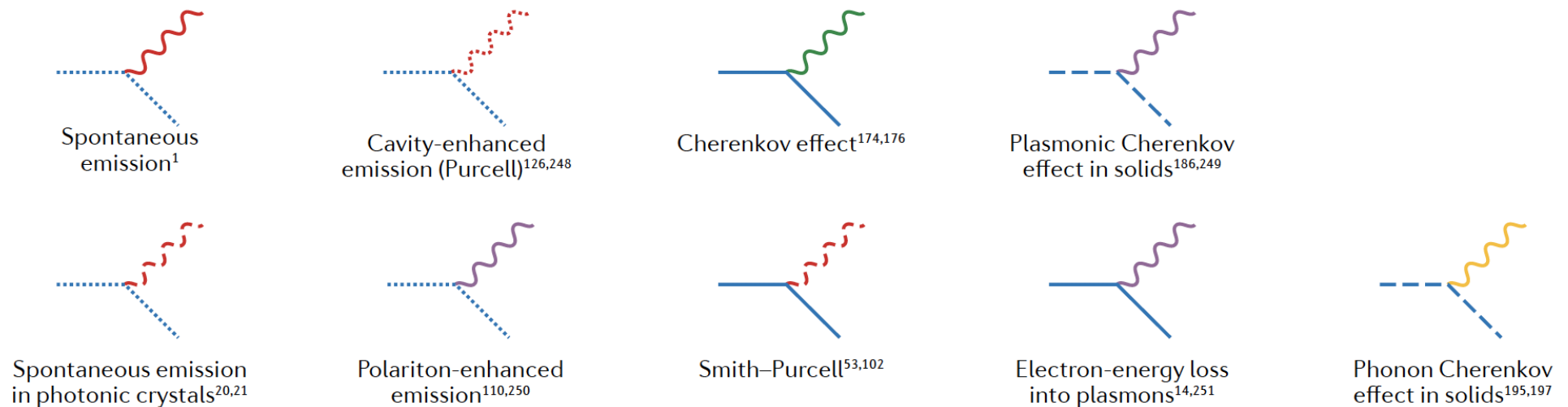
Elementary processes of light-matter interaction between electrons and photonic quasiparticles

Rivera*, Kaminer*, *et al. Science* (2016), Rivera *et al. Proc. Nat. Acad. Sci.* (2017),
Rivera *et al. Nature Physics* (2019), Rivera & Kaminer. *Nature Reviews Physics* (2020).

Describing light-matter interactions with photonic quasiparticles

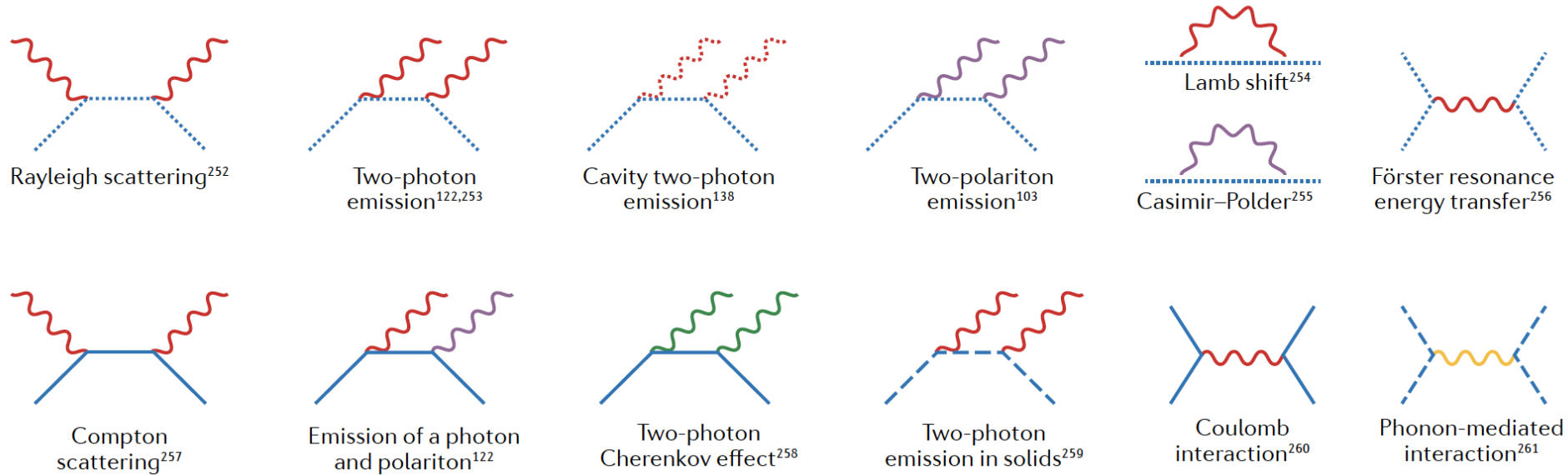


First-order processes described by MQED

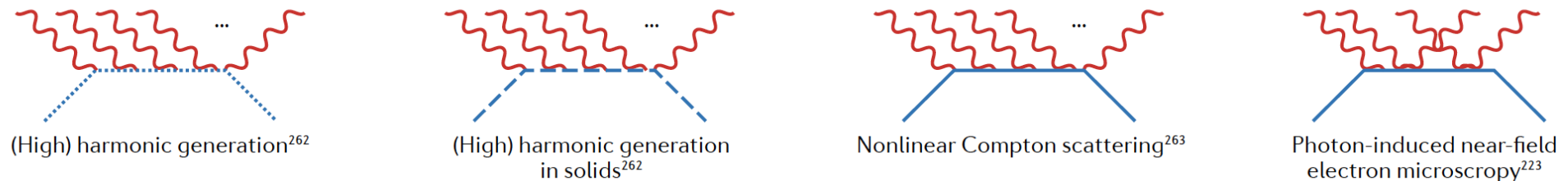


Describing light-matter interactions with photonic quasiparticles

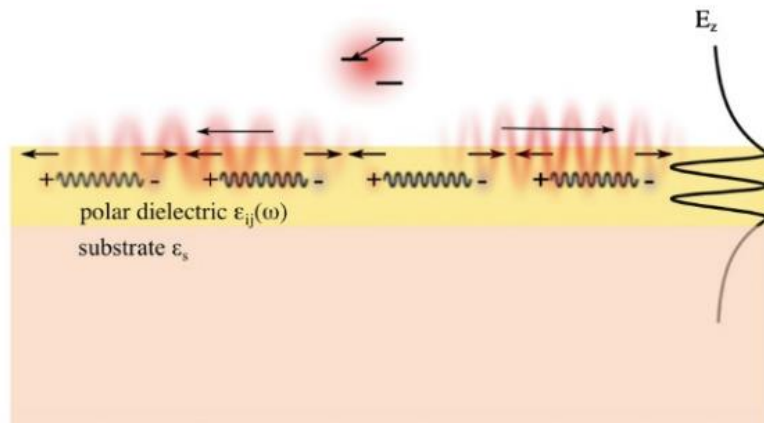
Second-order processes described by MQED



High-order processes by MQED



Higher-order processes can also be made dominant, like two-photon decay

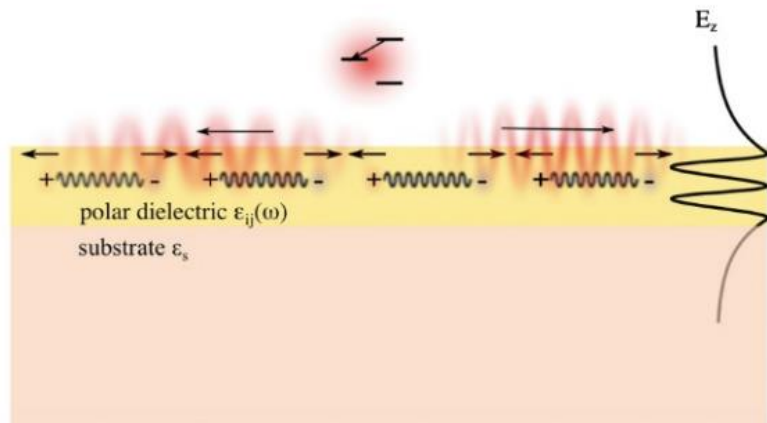


n -polariton emission rate:

$$\Gamma(nE1) \sim \alpha^n \eta^{3n} (ka)^{2n} \omega_0$$

Rivera *et al.* PNAS (2017).

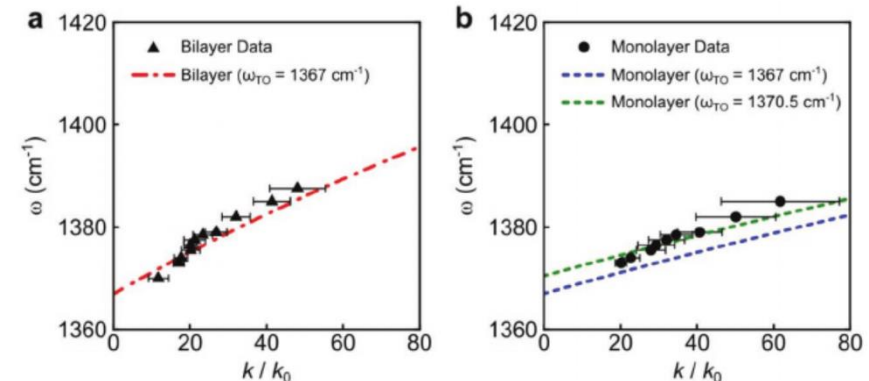
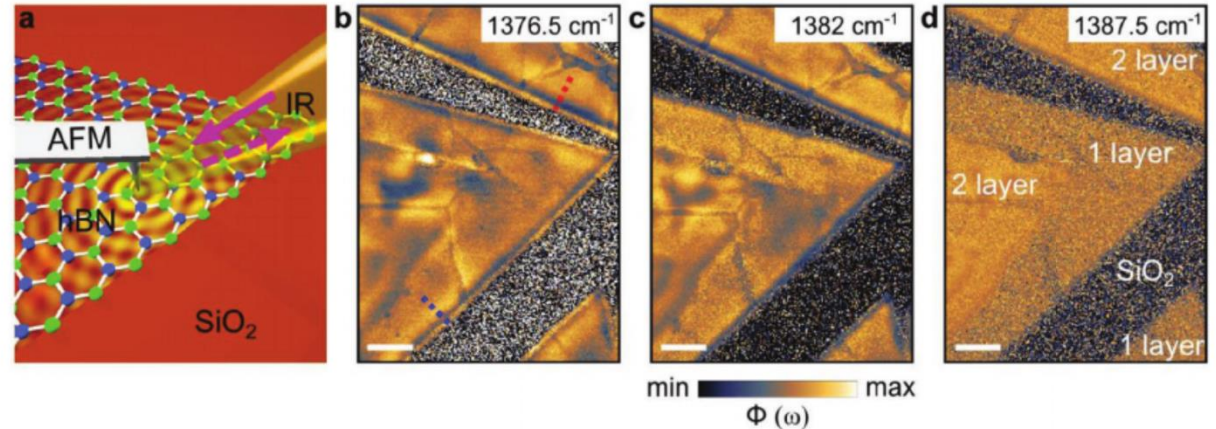
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n -polariton emission rate:

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Rivera *et al.* PNAS (2017).



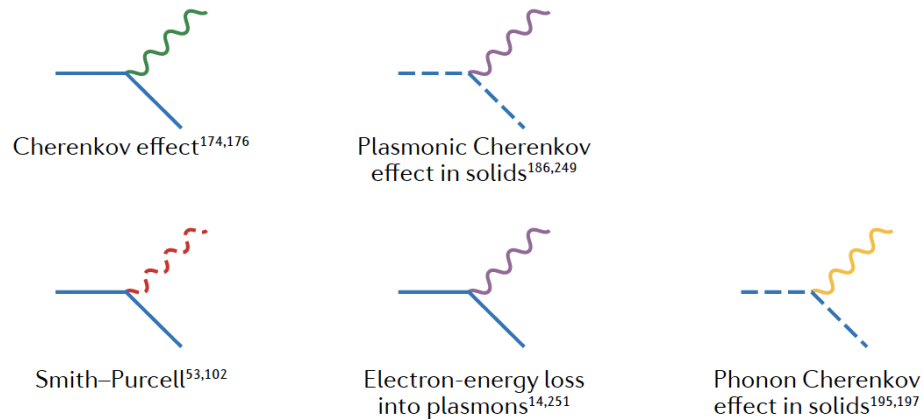
Predictions: Rivera *et al.* *Nano Lett.* (2019).

Observations: Dai, Fang, Rivera, *et al.* *Adv. Mat.* (2019).

Li *et al.* *Nature Materials* (2021).

[collabs. with P. Narang (Harvard), D. Basov (Columbia)]

New physics in free-electron light-matter interactions with photonic quasiparticles

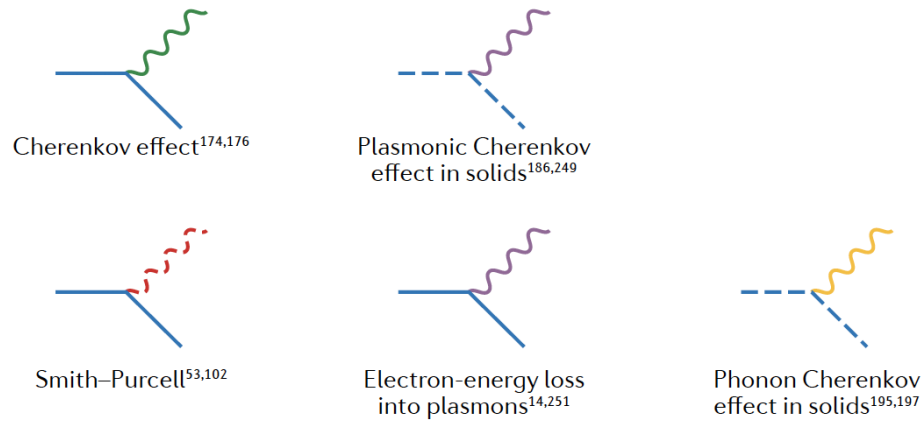


Rivera & Kaminer, *Nat. Rev. Phys.* (2020)

[for even more, see free-electron quantum optics collaboration with Prof. Ido Kaminer (Technion)]

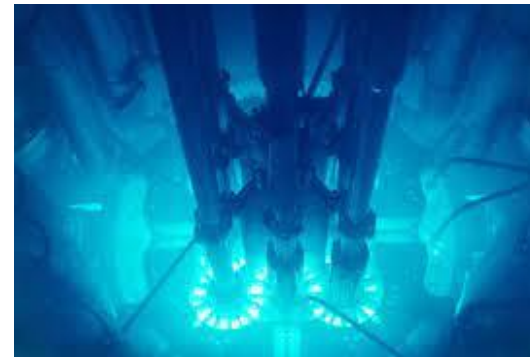
New physics in free-electron light-matter interactions with photonic quasiparticles

Cherenkov radiation, with and without PQPs



Rivera & Kammer, *Nat. Rev. Phys.* (2020)

[for even more, see free-electron quantum optics collaboration with Prof. Ido Kammer (Technion)]

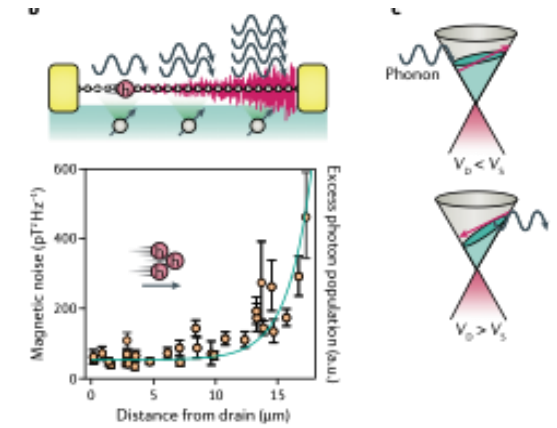


Cherenkov effect

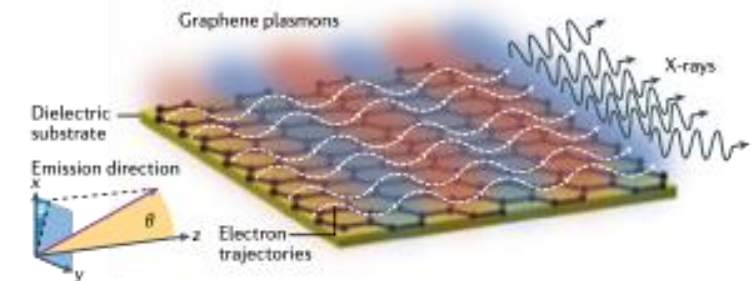
Free-electron X-ray sources, with and without PQPs



Synchrotron

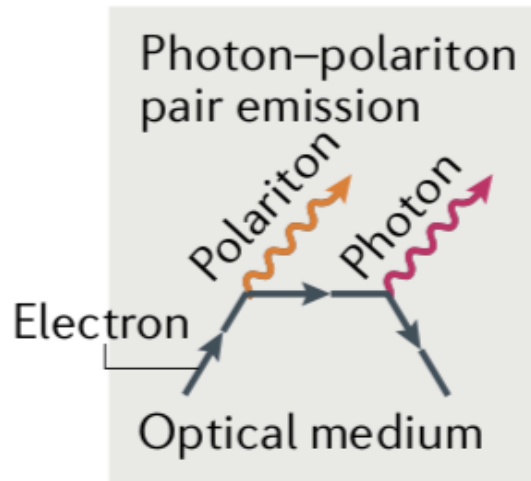


Andersen *et al.*, *Science* (2019)



Wong *et al.*, *Nature Photonics* (2016)

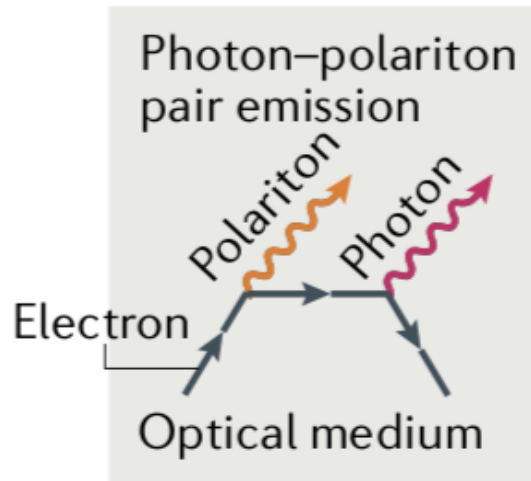
Two-photon emission by a free electron into an X-ray and a polariton



$$\omega_{\text{ph}} = \omega_{\text{pol}} \frac{\beta n(\omega_{\text{pol}}) \cos \theta_{\text{pol}} - 1}{1 - \beta \cos \theta_{\text{ph}}}$$

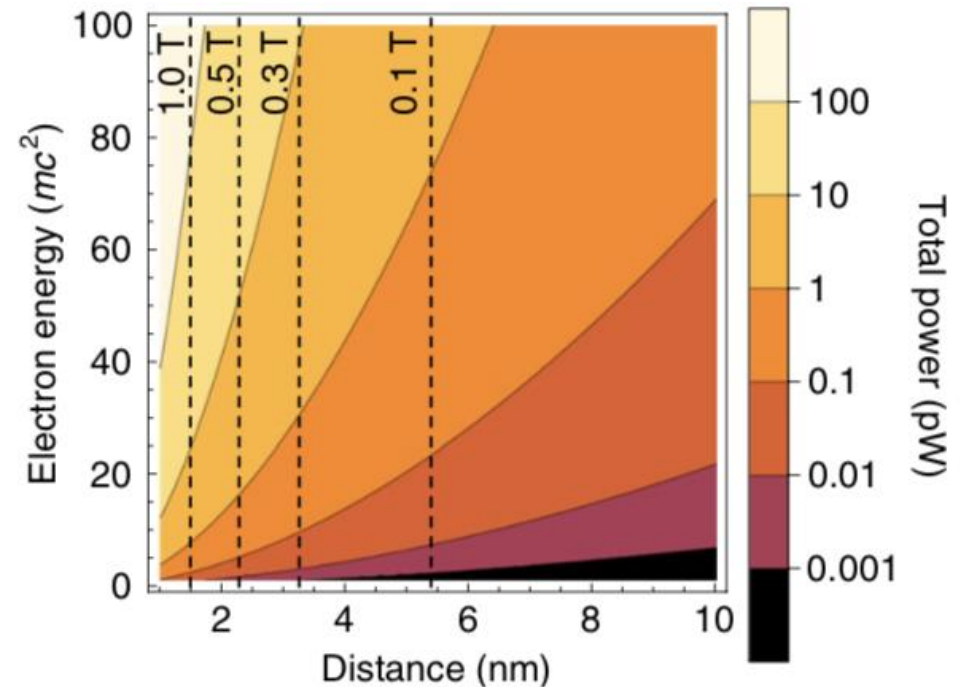
Enables simultaneous emission of an (entangled) polariton/X-ray photon pair

Two-photon emission by a free electron into an X-ray and a polariton



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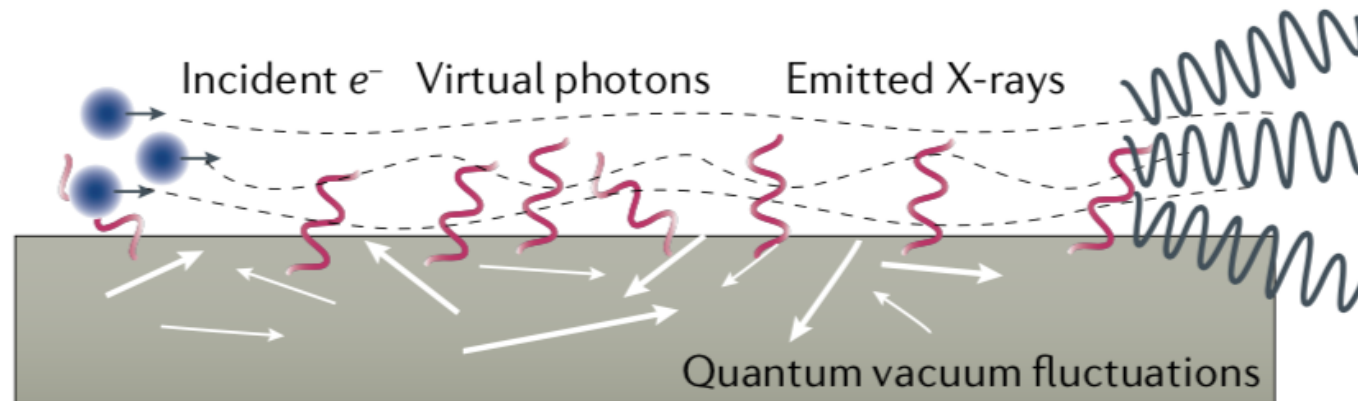
Enables simultaneous emission of an (entangled) polariton/X-ray photon pair



Power comparable to synchrotron radiation in a Tesla-scale field

The two-photon emission is strong, due to strong nanoscale vacuum forces

$$P = \frac{e^4 \gamma^2 (4 - \beta^2)}{24\pi \epsilon_0 m^2 c^3} \langle 0 | \mathbf{E}^2 | 0 \rangle \quad \text{[Larmor's formula]}$$



This TPE can be equivalently calculated as radiation by an accelerated charge, which comes from a vacuum (Casimir-like) fluctuating force from zero-point fluctuations of the photonic quasiparticle (here, plasmons).

New particle detectors (scintillators) based on photonic quasiparticle interactions

Roques-Carmes*, Rivera* *et al.* *A general framework for scintillation in nanophotonics.* arXiv: 2110.11492.



Charles Roques-Carmes

Scintillators: an application of light-matter interactions with photonic quasiparticles

Core phenomenon

Ionizing radiation
(e^- , X , γ)

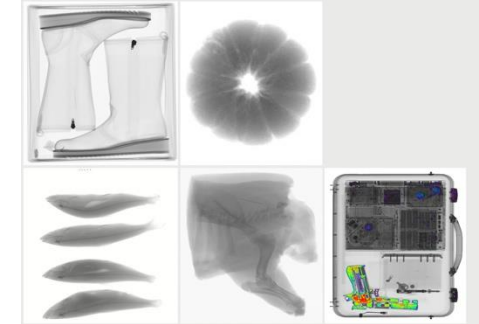


Application domains

Medical imaging



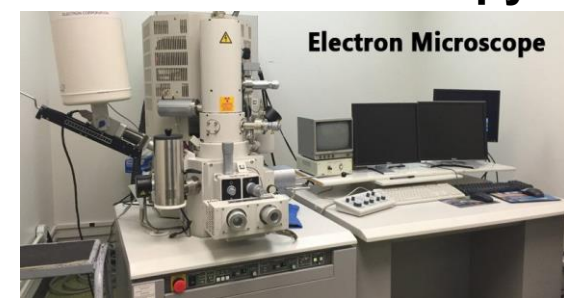
Non-destructive testing



Radiation monitoring

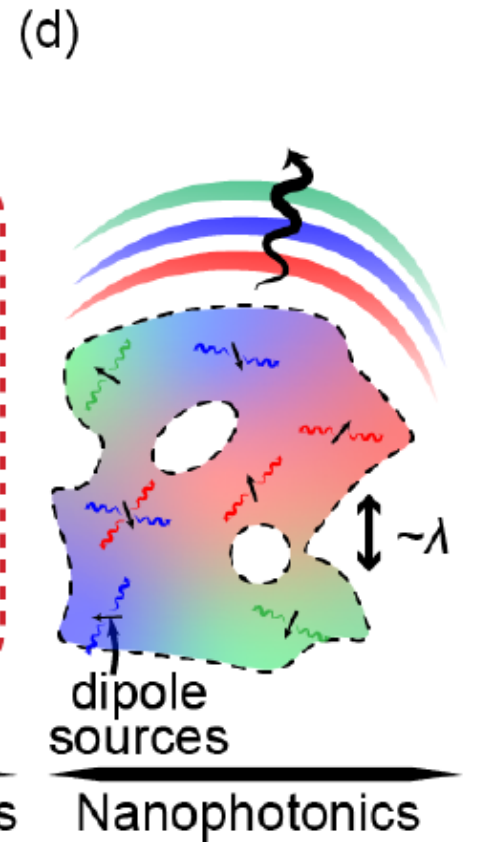
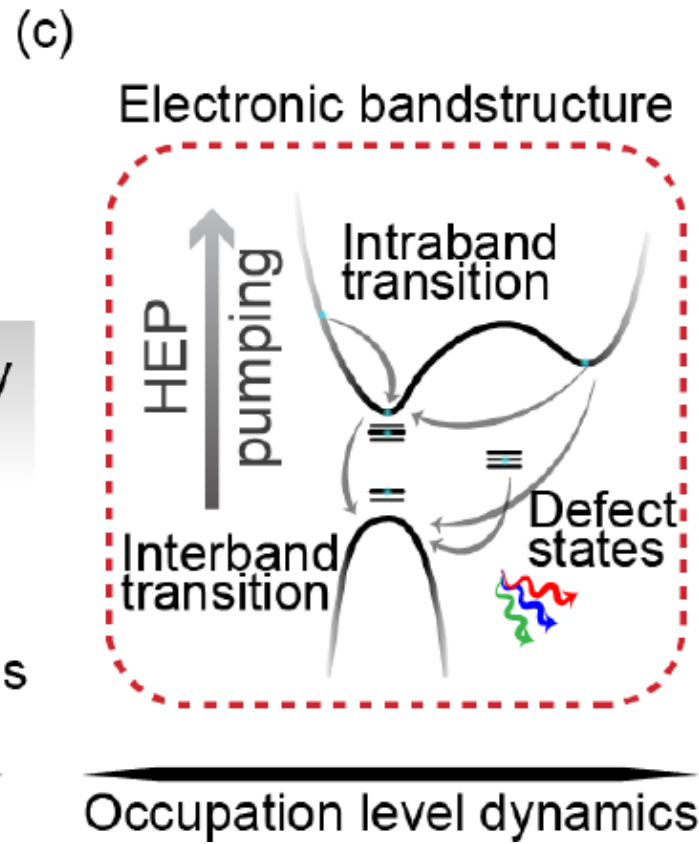
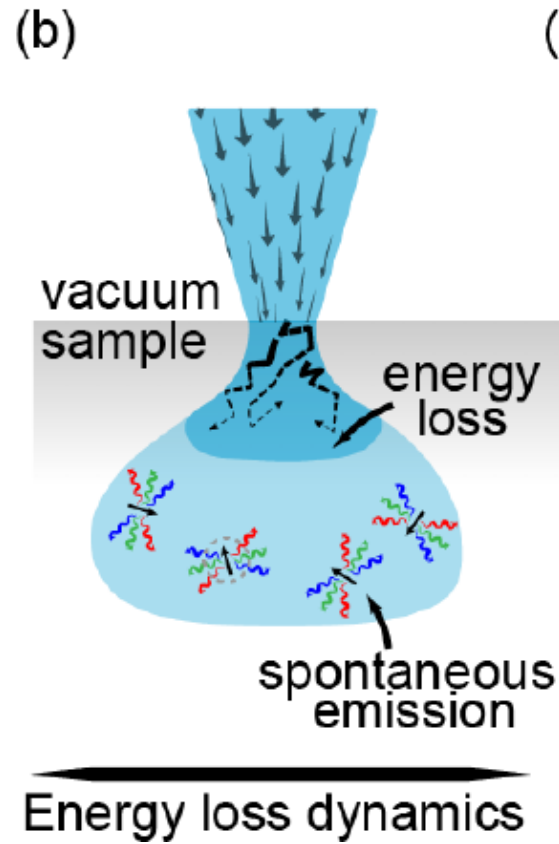
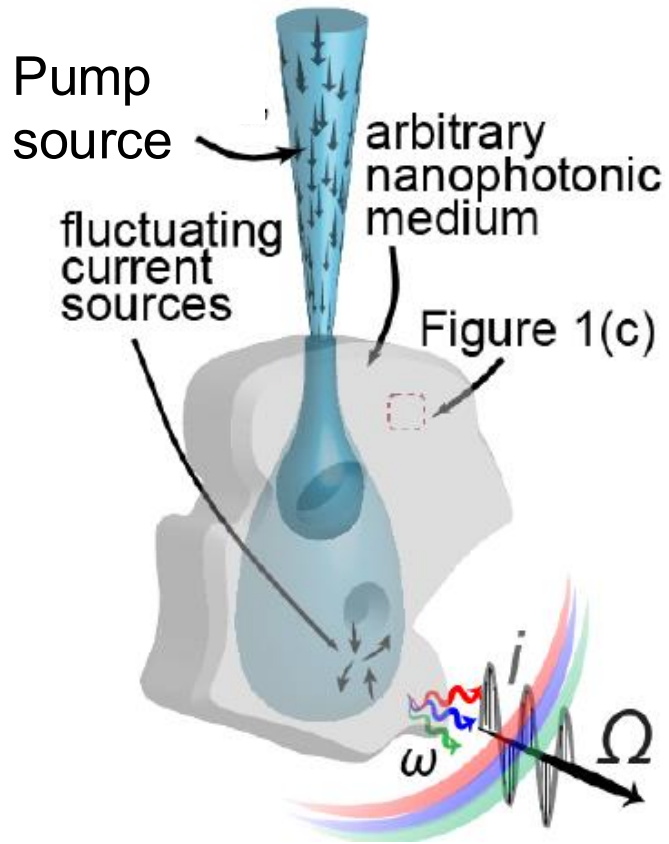


Electron microscopy

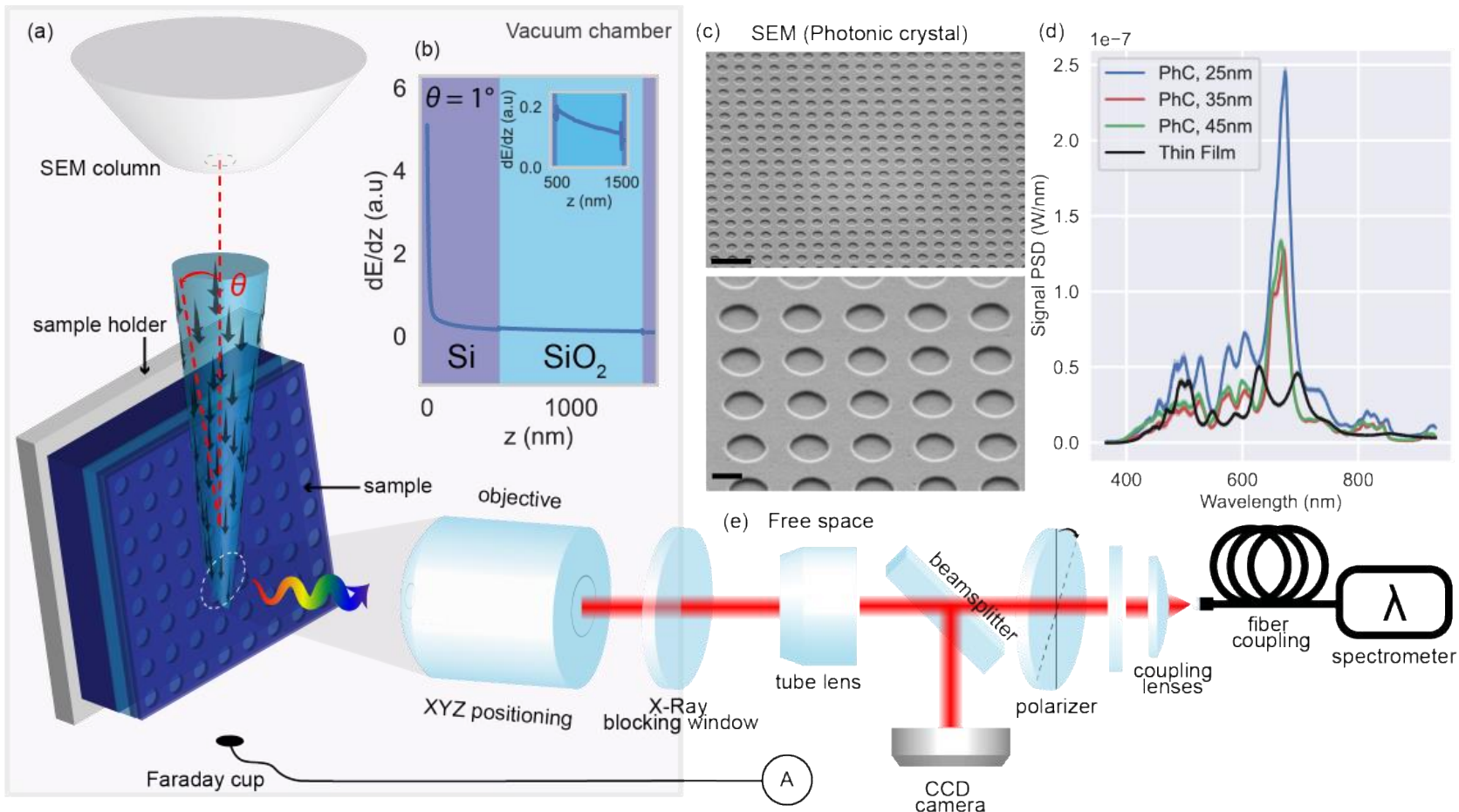


A tantalizing prospect: enhancing scintillation brightness with nanophotonic control over the density of states – but it's complicated!

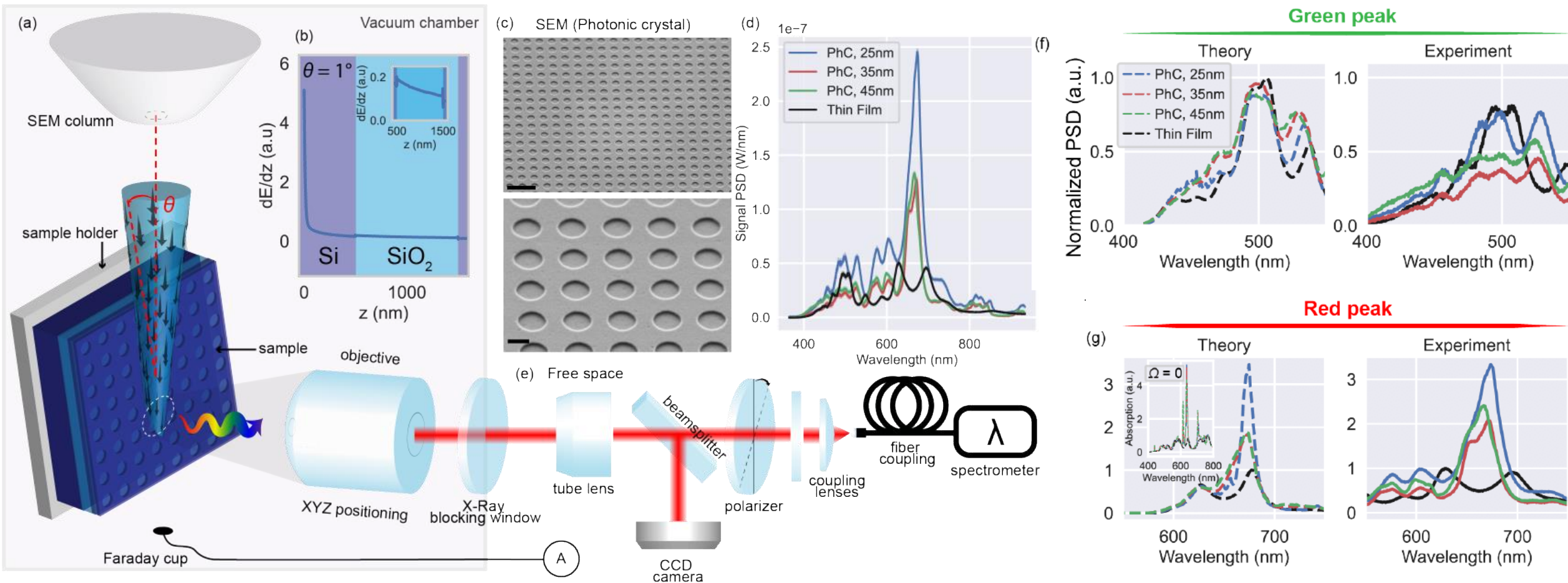
The process of scintillation, from end-to-end



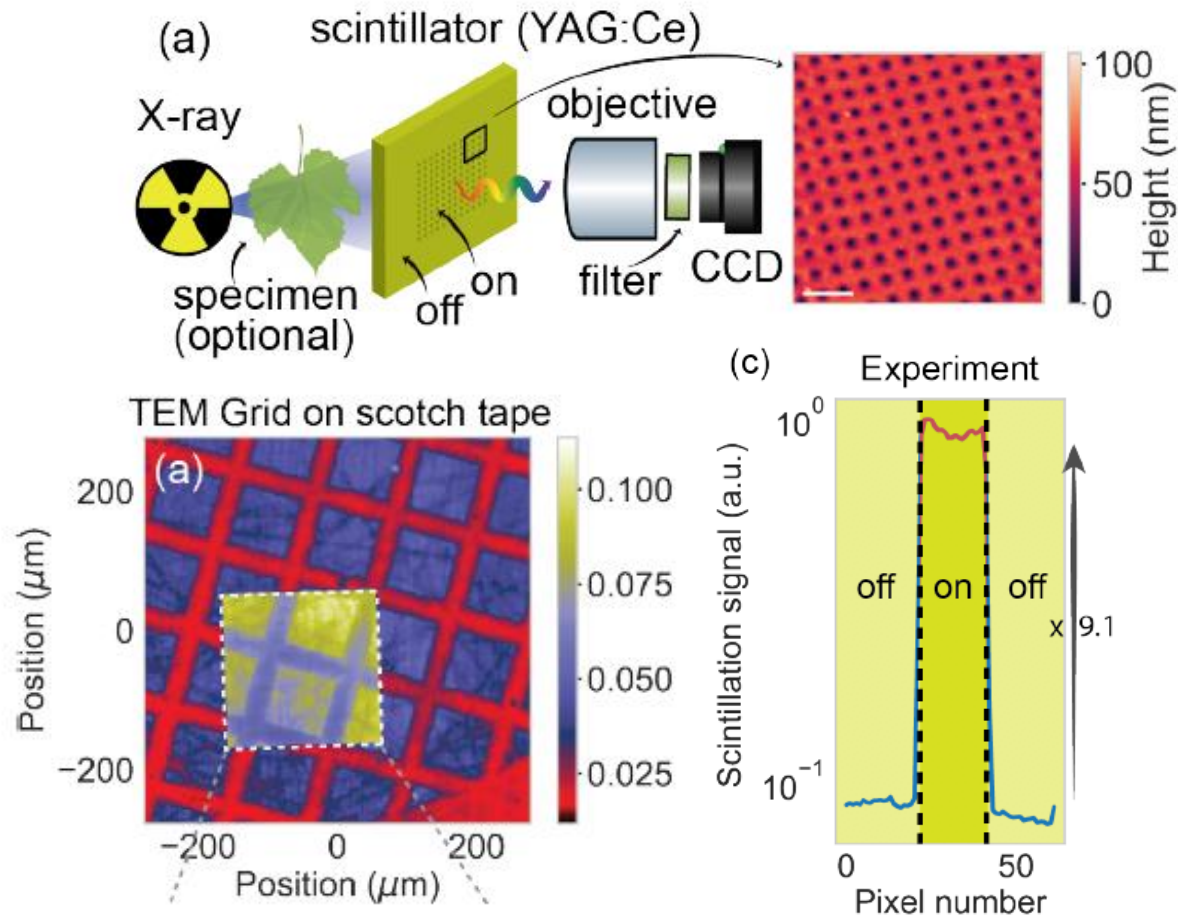
Nanophotonic-enhanced scintillation induced by electron bombardment



Nanophotonic-enhanced scintillation induced by electron bombardment



Nanophotonic-enhanced scintillation induced by X-ray bombardment

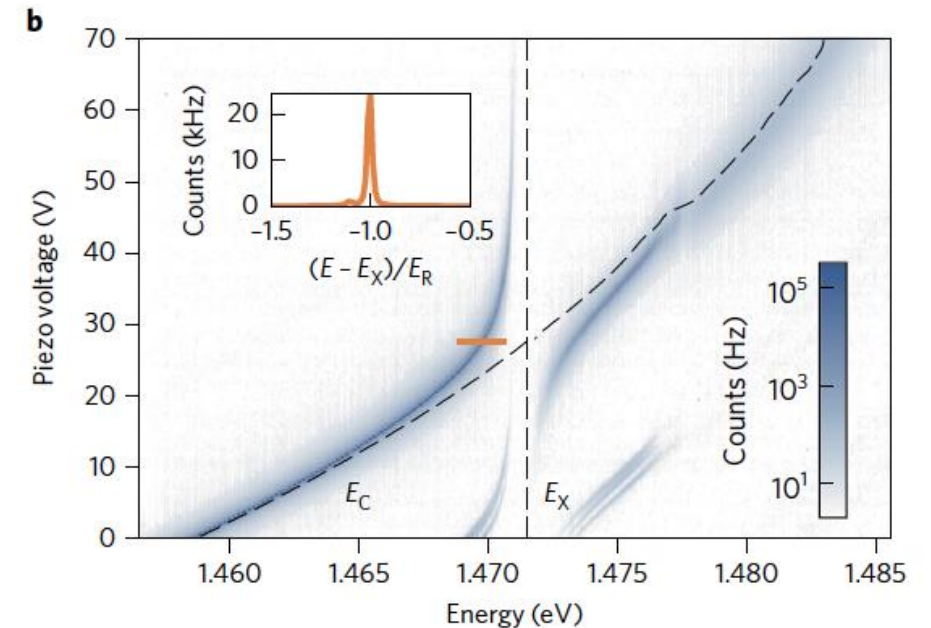
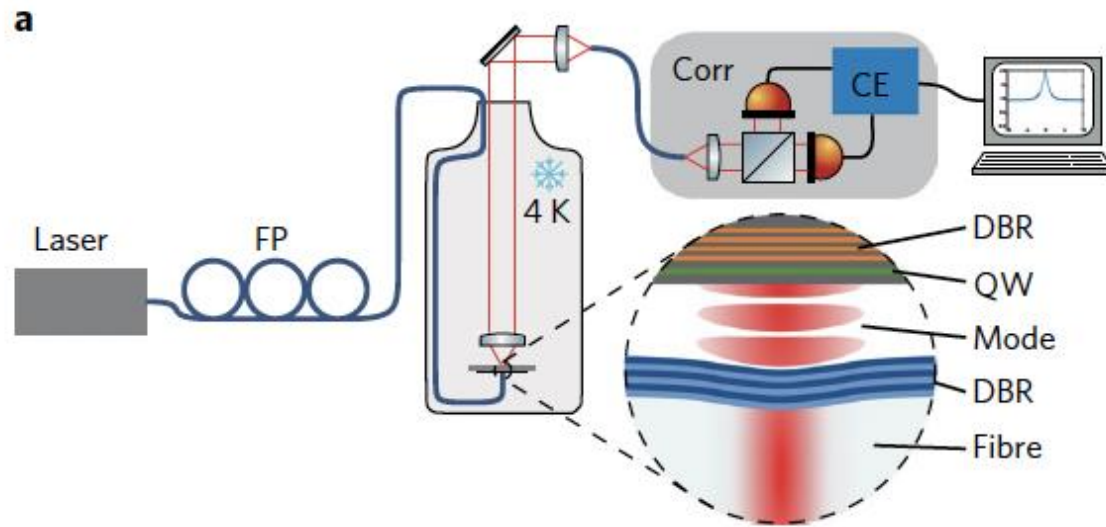


- Real-world example
 - Patterned photonic crystal into an X-ray scintillator
 - Put it into a micro-CT machine and look at light from patterned vs. unpatterned region
- **10x brighter image** from the patterned region

Optical nonlinearities and preparation of macroscopic optical Fock states

Rivera et al. Macroscopic condensation of photon noise in sharply nonlinear dissipative systems. In preparation.

Photonic quasiparticles can also have (much) stronger optical nonlinearities



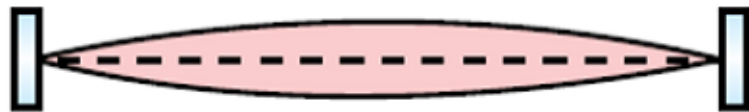
Such systems realize a Kerr Hamiltonian

$$H_{\text{Kerr}}/\hbar = \omega_{\text{LP}}(p^\dagger p + \beta p^{\dagger 2} p^2)$$

where $\beta \sim 10^{-5}$. Compare to expected value $\chi^{(3)}(\pi\hbar c/\epsilon_0\lambda^4) \lesssim 10^{-8}$ (GaAs).

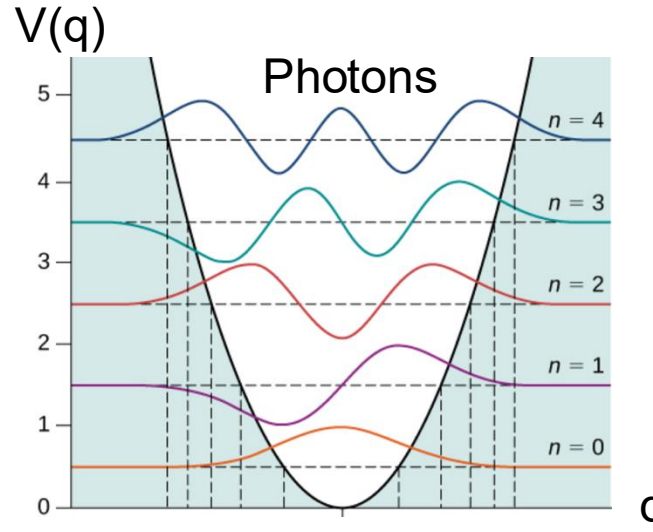
Dissipative Kerr physics in exciton-polaritons
 Imamoglu *et al.* Physical Review Letters (1997).
 Fink *et al.* Nature Physics (2018).
 Delteil & Fink *et al.* Nature Materials (2019).

Using strong nonlinearities to generate macroscopic Fock states of radiation



$$A(z, t) = 2\sqrt{\frac{\hbar\omega}{\epsilon_0 V}} \sin(kz)q$$

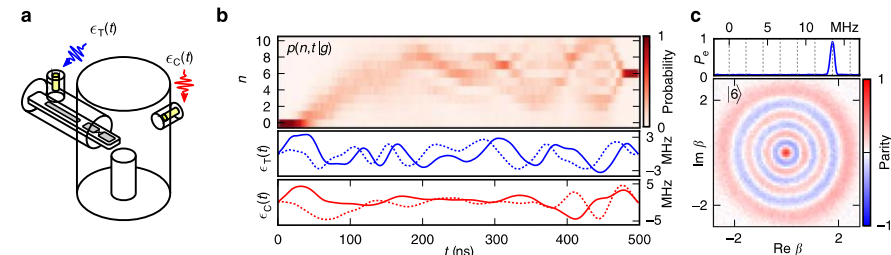
$$a^\dagger a |n\rangle = n |n\rangle$$



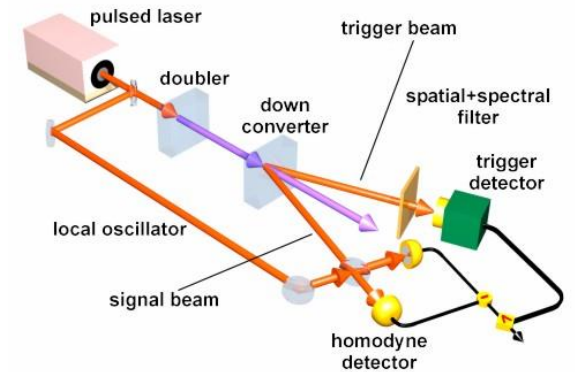
Fock states are the most fundamental states of light, and yet extremely difficult to produce

- Few mechanisms naturally selecting Fock
- Fragile in presence of loss

How Fock states are generated



Heeres *et al. Nat. Comm.* (2017).



Lvovsky, PRL (2001),
Bimbard *Nat. Phot.* (2010).

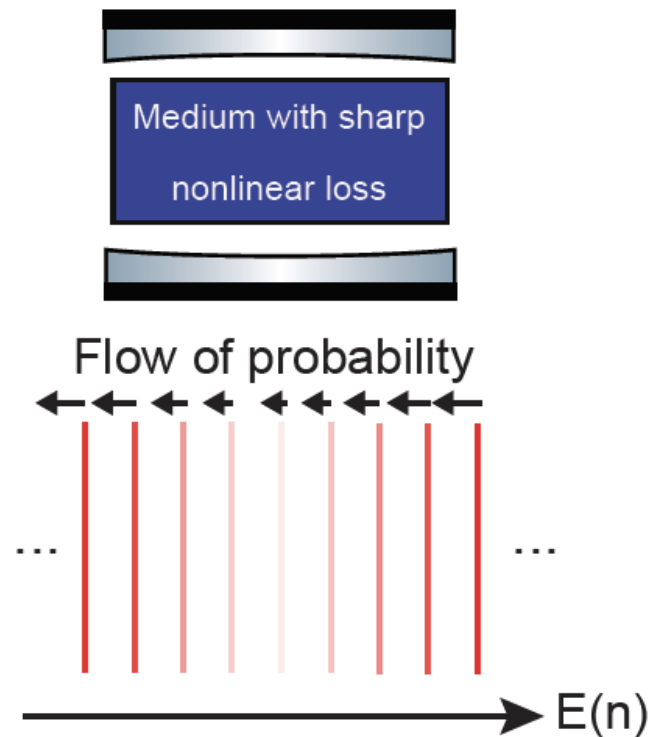
Two steps to producing macroscopic Fock states of radiation

- (1) Construct a special nonlinear (intensity-dependent) dissipation
 - Time-dependent solution of master equation: quantum light states will evolve into a Fock state **transiently** (or close, sub-Poissonian approximation)

- (2) Incorporate a gain medium into (1) [now it's a laser]
 - **Steady-state** solution of master equation: Fock state or close approximation

Dissipative preparation of large Fock states

Starting point: decay of light in a cavity with an embedded nonlinear medium.
The effect of the NL medium will be to induce an intensity-dependent dissipation.



Effect of decay on the photon probabilities?

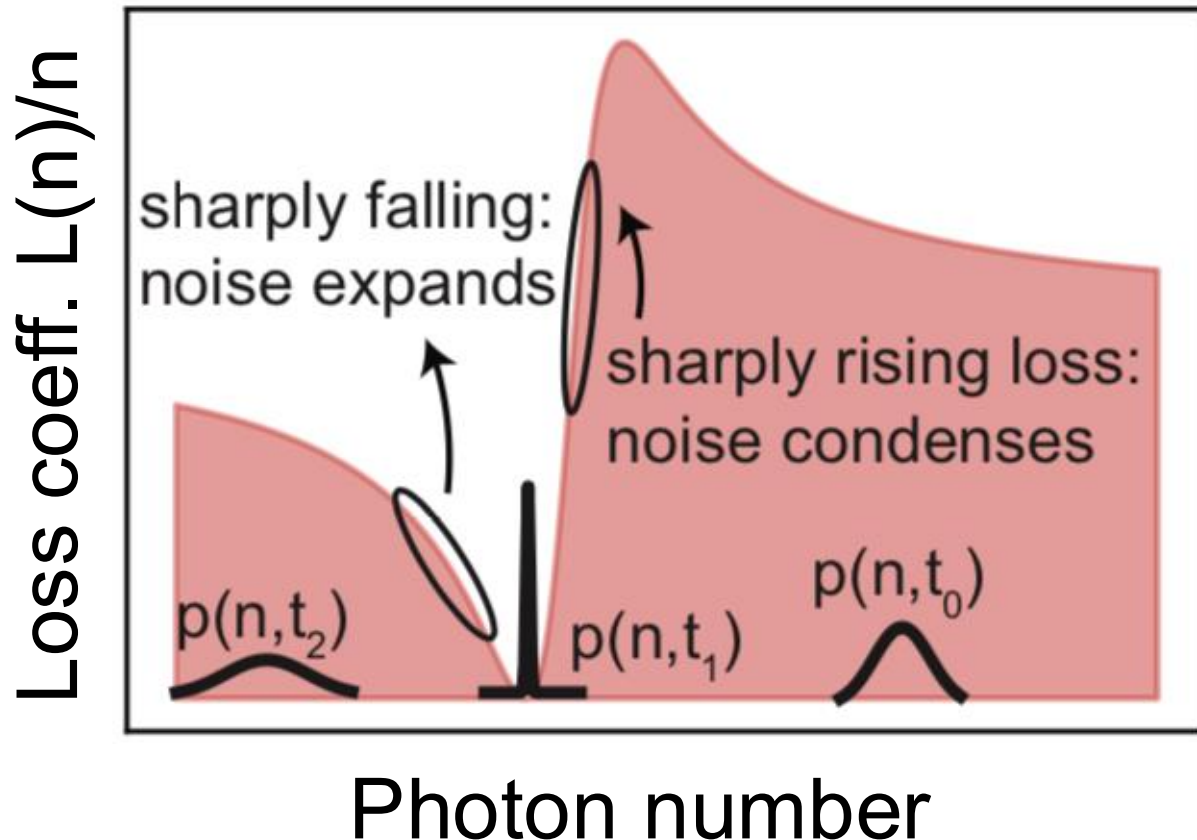
$$\dot{p}(n) = -L(n)p(n) + L(n+1)p(n+1)$$

Rate of transition in which n photons becomes $n-1$

Rate of transition in which $n+1$ photons becomes n

[master equation for probabilities]

A nonlinear interaction that naturally produces Fock states of light



Dynamics of first two cumulants:

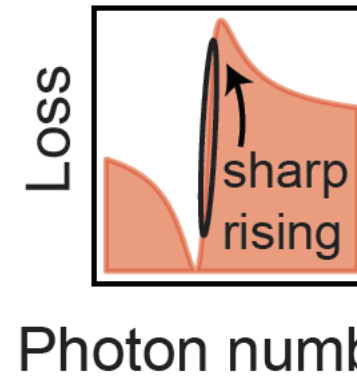
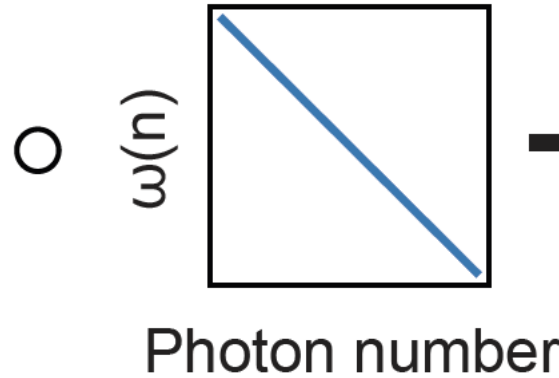
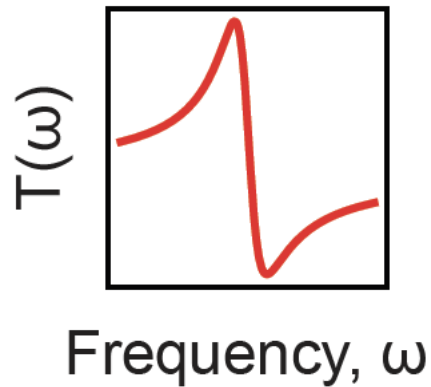
$$\dot{n} = -L(n)$$

$$(\Delta \dot{n})^2 = L(n) - 2(dL/dn)(\Delta n)^2$$

Photon noise drops relative to mean:

$$dL/dn(\Delta n)^2 > L(n)$$

How to create such a nonlinear loss?



$$\frac{d\kappa}{dn} = \frac{d\kappa}{d\omega} \frac{d\omega}{dn} \sim \beta/\gamma$$

[Sharper transmission & stronger NL, yield stronger effect]

A physical picture of what's happening:

- As light leaks out, intensity changes
- As intensity changes, index & mode freq. changes [Kerr effect]
- Variation of mode frequency relative to mirror freq. changes loss

An explicit construction of nonlinear loss

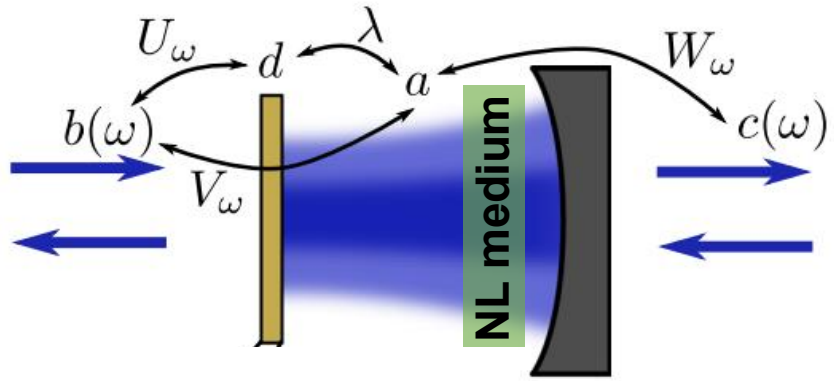


Image source: Cernotik *et al.* PRL (2019).

$$H/\hbar = H_a + \omega_d d^\dagger d + (\lambda a d^\dagger + \lambda^* a^\dagger d) + \sum_k \omega_k b_k^\dagger b_k \\ + \sum_k (g_k a b_k^\dagger + g_k^* a^\dagger b_k) + \sum_k (v_k d b_k^\dagger + v_k^* d^\dagger b_k).$$

Rivera *et al.* In preparation.

An explicit construction of nonlinear loss

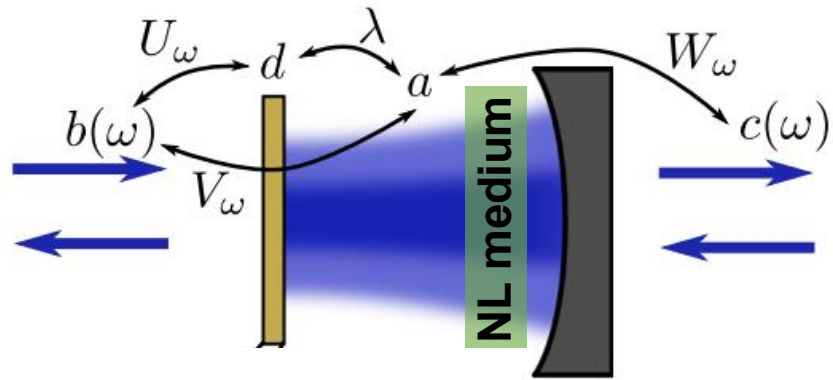


Image source: Cernotik *et al.* PRL (2019).

$$H/\hbar = H_a + \omega_d d^\dagger d + (\lambda a d^\dagger + \lambda^* a^\dagger d) + \sum_k \omega_k b_k^\dagger b_k + \sum_k (g_k a b_k^\dagger + g_k^* a^\dagger b_k) + \sum_k (v_k d b_k^\dagger + v_k^* d^\dagger b_k).$$

Rivera *et al.* In preparation.

Master equation for an anharmonic oscillator coupled to a frequency-dependent loss.

$$\dot{\rho}_{ad} = -i[H_{ad}/\hbar, \rho_{ad}] + (X^\dagger X \rho_{ad} + \rho_{ad} X^\dagger X - 2X \rho_{ad} X^\dagger).$$

$$X = \sqrt{\kappa} a + \sqrt{\gamma} d$$

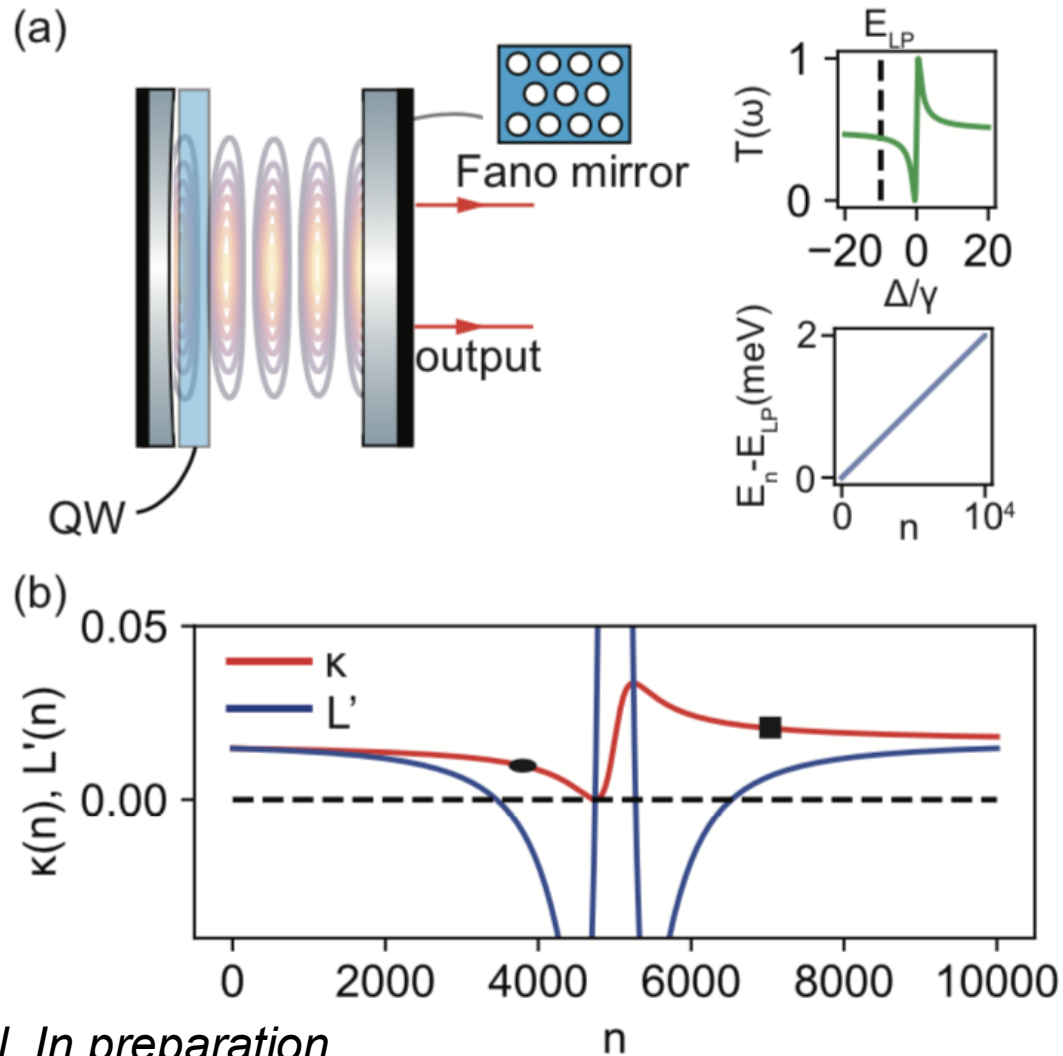


adiabatic elimination of mirror mode (d)

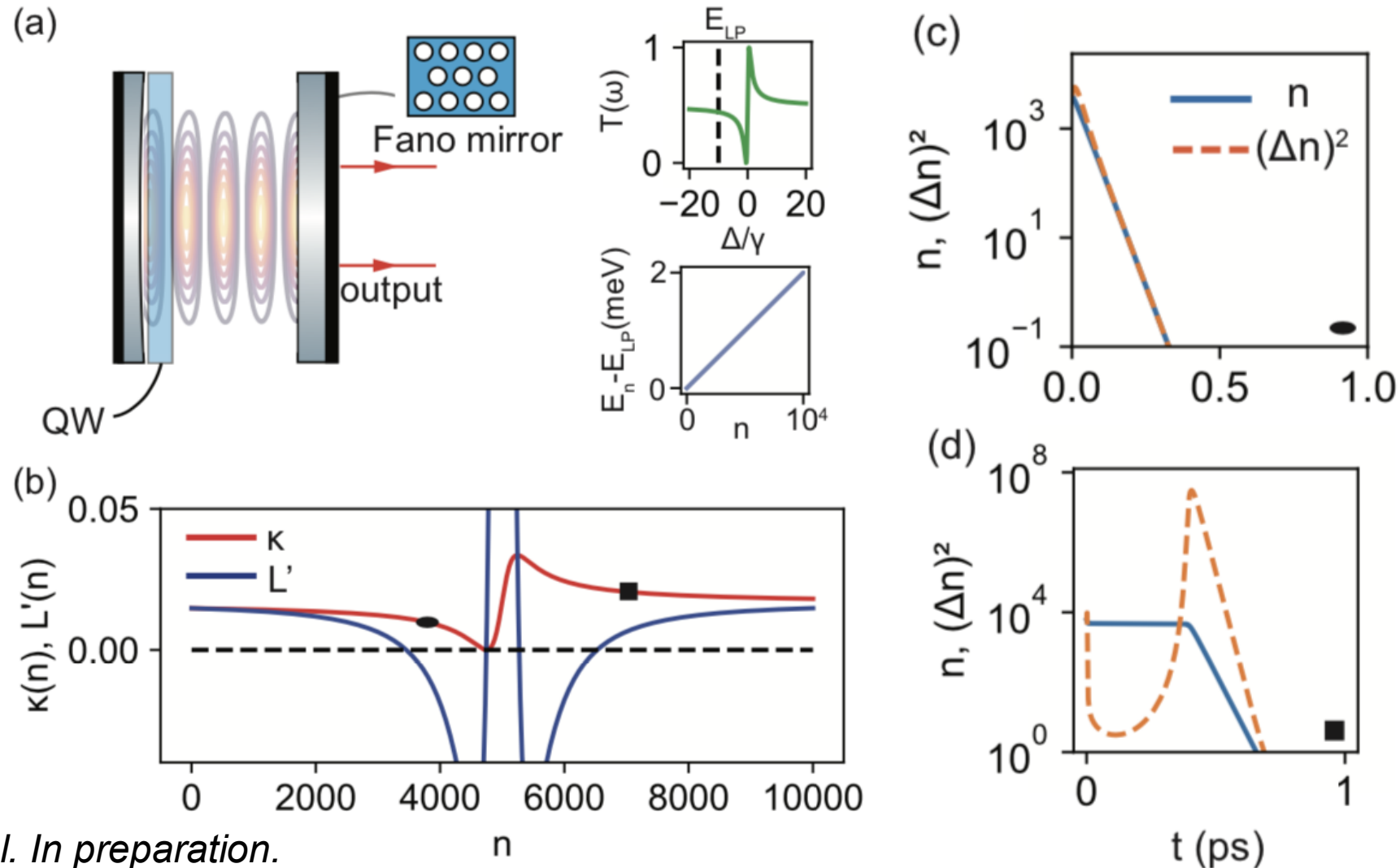
$$\dot{\rho}_{n,n} = -n \left(\kappa - 2\text{Re} \left[\frac{G_+ G_-}{i(\omega_d - \omega_{n,n-1}) + \gamma/2} \right] \right) \rho_{n,n} + (n+1) \left(\kappa - 2\text{Re} \left[\frac{G_+ G_-}{i(\omega_d - \omega_{n+1,n}) + \gamma/2} \right] \right) \rho_{n+1,n+1},$$

$$L(n+1)$$

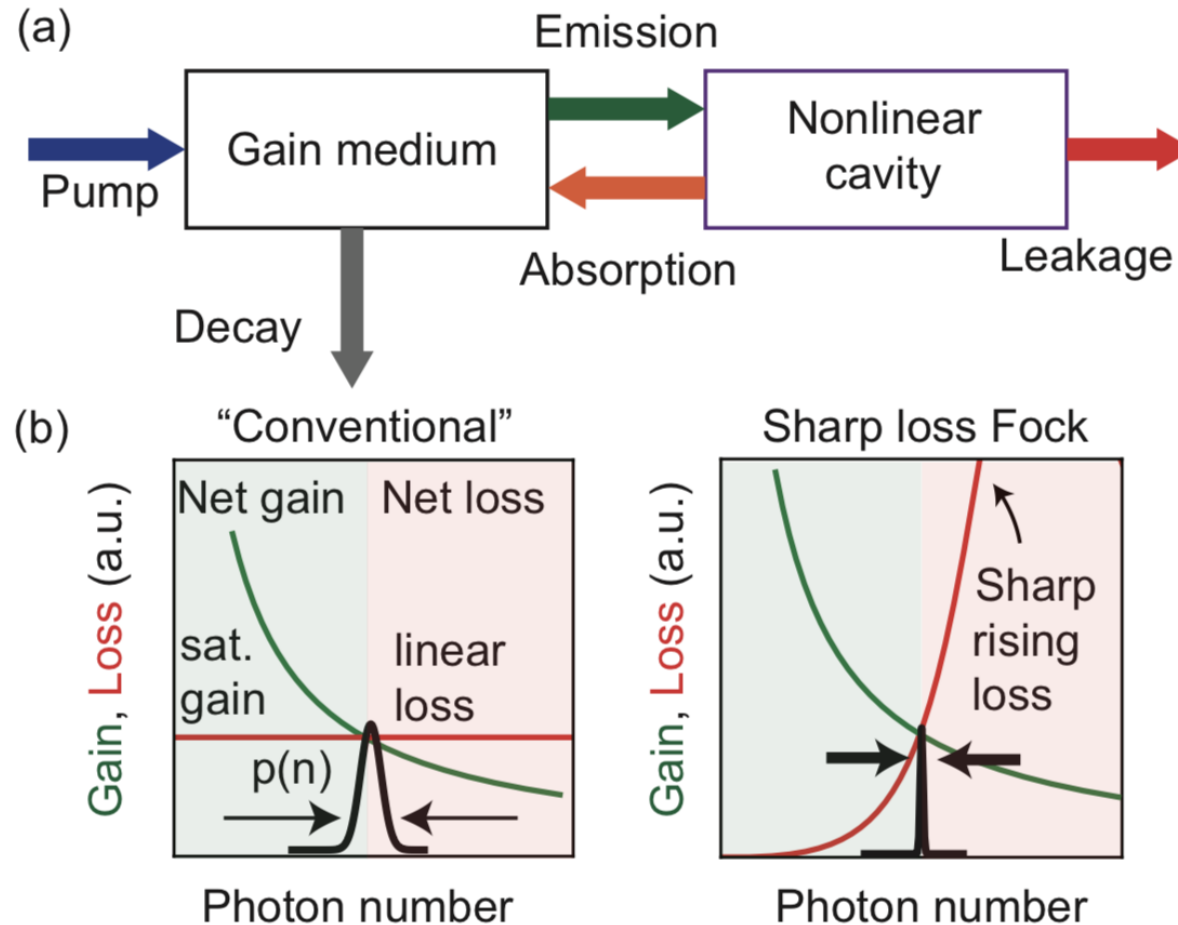
Nearly complete condensation of optical photon noise using this nonlinearity



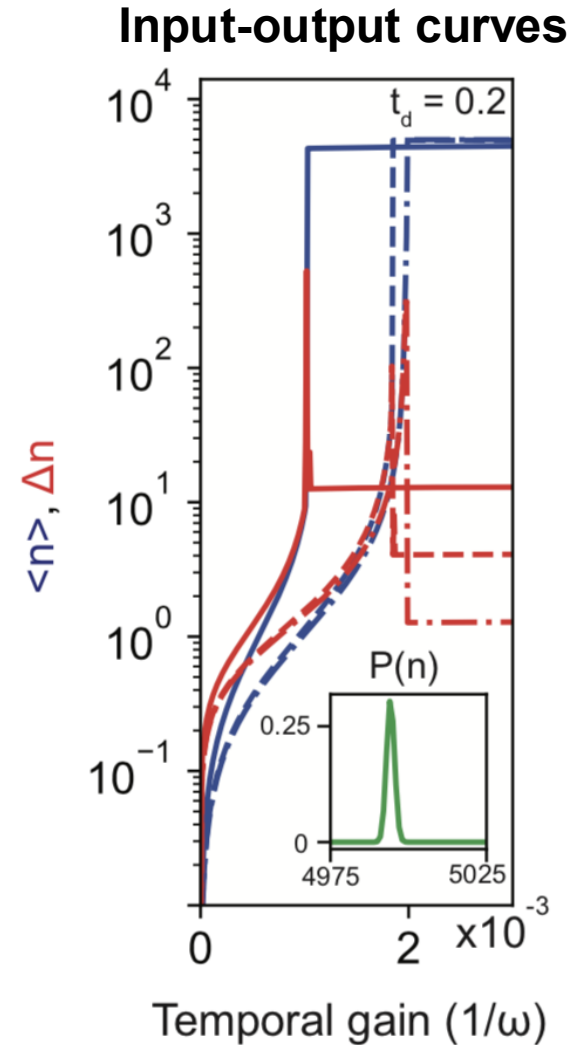
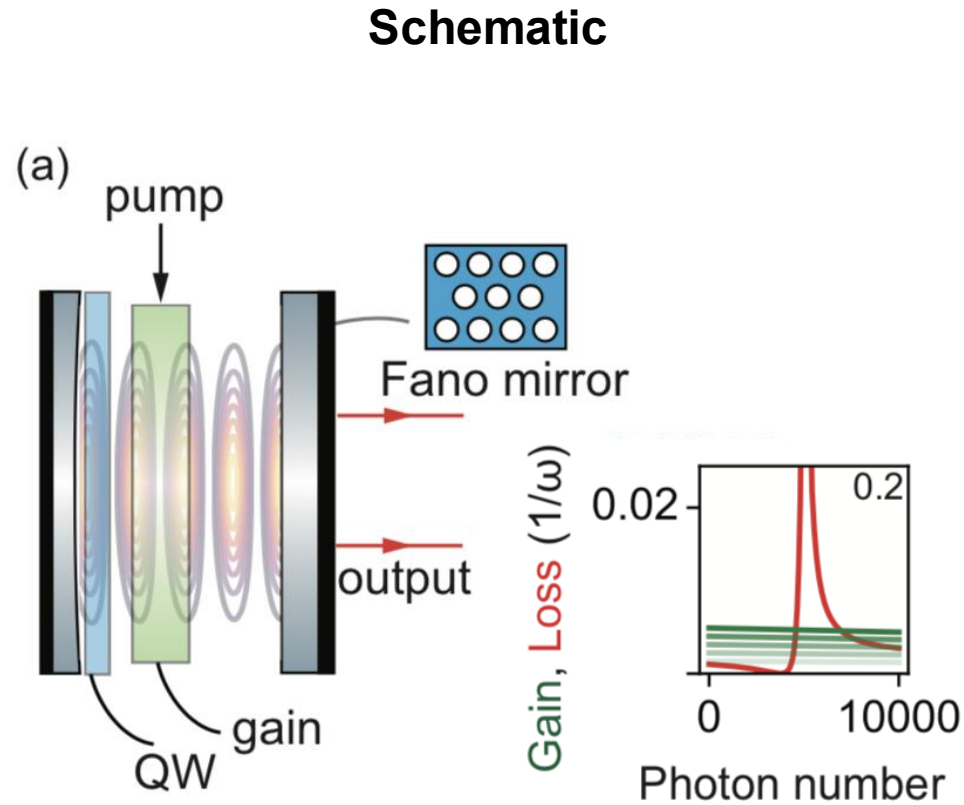
Nearly complete condensation of optical photon noise using this nonlinearity



This nonlinear interaction enables a new kind of laser emitting Fock states



Characteristics of an optical Fock laser



**Condensation of optical noise in the
deeply macroscopic regime?**

Acknowledgements

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Jamison Sloan (MIT PhD EECS),

Ali Ghorashi (MIT PhD physics)

Yannick Salamin (MIT postdoc)



Supplementary slides

Macroscopic QED as a technique to study light-matter coupling in dissipative systems

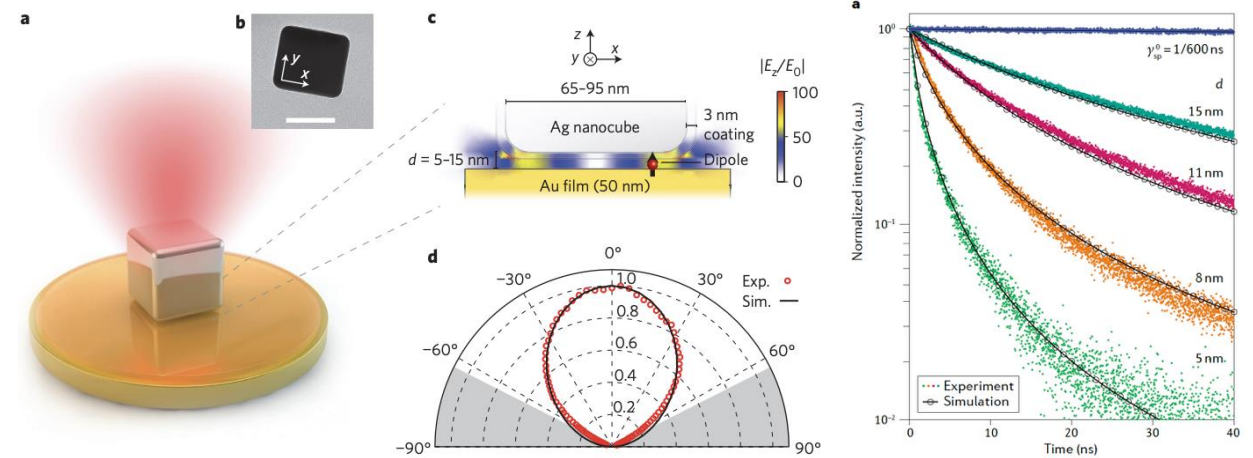
Rivera*, Kaminer*, *et al. Science* (2016), Rivera *et al. Proc. Nat. Acad. Sci.* (2017),
Rivera *et al. Nature Physics* (2019), Rivera & Kaminer. *Nature Reviews Physics* (2020).

Confining light on wavelength scales to control light-matter interaction

- Many of the applications above: light is a *plane wave*, with energy delocalized over a volume $\gg \lambda^3$.
- **The interactions are *very weak*, and are largely lowest-order (e.g., in coupling strength, atomic size, field strength)**
- A promising “way out” is nanophotonics
 - Controlling & confining light with polarizable media with spatial variations on the scale of λ (*nanophotonics*), we can enhance the interactions.
 - Intuitively: changing basic properties of the photon (soln. to Maxwell)

Confining light on wavelength scales to control light-matter interaction

- Many of the applications above: light is a *plane wave*, with energy delocalized over a volume $\gg \lambda^3$.
- By controlling light with polarizable media with spatial variations on the scale of λ (*nanophotonics*), we can enhance the interactions.
- Very representative: Purcell effect
- **Still, the interactions are very weak, and are largely lowest-order (e.g., in coupling strength, atomic size, field strength)**



Akselrod *et al.* *Nature Photonics* (2014).

$$\Gamma_{fi} = \frac{3}{4\pi^2} \frac{Q}{(V/\lambda_0^3)} |\hat{\mathbf{d}}_{fi} \cdot \mathbf{u}(\mathbf{r})|^2 \Gamma_0,$$

Relation to textbook EM field quantization

Table 1 | Levels of quantization of the electromagnetic field

| Type of medium | Vector potential | Conditions on vector potential |
|--|---|---|
| Vacuum | $\sum_{\mathbf{k}, \epsilon_{\mathbf{k}}} \sqrt{\frac{\hbar}{2\epsilon_0\omega_{\mathbf{k}}V}} \left(e^{i\mathbf{k}\cdot\mathbf{r}-i\omega_{\mathbf{k}}t} \epsilon_{\mathbf{k}} a_{\mathbf{k}, \epsilon_{\mathbf{k}}} + e^{-i\mathbf{k}\cdot\mathbf{r}+i\omega_{\mathbf{k}}t} \epsilon_{\mathbf{k}}^* a_{\mathbf{k}, \epsilon_{\mathbf{k}}}^\dagger \right)$ | $\mathbf{k} \cdot \epsilon_{\mathbf{k}} = 0; \omega_{\mathbf{k}} = ck$ |
| Homogeneous, lossless, non-dispersive, isotropic | $\sum_{\mathbf{k}, \epsilon_{\mathbf{k}}} \sqrt{\frac{\hbar}{2\epsilon_0\omega_{\mathbf{k}}n^2V}} \left(e^{i\mathbf{k}\cdot\mathbf{r}-i\omega_{\mathbf{k}}t} \epsilon_{\mathbf{k}} a_{\mathbf{k}, \epsilon_{\mathbf{k}}} + e^{-i\mathbf{k}\cdot\mathbf{r}+i\omega_{\mathbf{k}}t} \epsilon_{\mathbf{k}}^* a_{\mathbf{k}, \epsilon_{\mathbf{k}}}^\dagger \right)$ | $\mathbf{k} \cdot \epsilon_{\mathbf{k}} = 0; \omega_{\mathbf{k}} = ck/n$ |
| Inhomogeneous, lossless, non-dispersive, anisotropic | $\sum_n \sqrt{\frac{\hbar}{2\epsilon_0\omega_n}} \left(\mathbf{F}_n(\mathbf{r}) e^{-i\omega_n t} a_n + \mathbf{F}_n^*(\mathbf{r}) e^{i\omega_n t} a_n^\dagger \right)$ | $\nabla \times \nabla \times \mathbf{F}_n(\mathbf{r}) = \boldsymbol{\epsilon}(\mathbf{r}) \frac{\omega_n^2}{c^2} \mathbf{F}_n(\mathbf{r});$ $\int d^3r \mathbf{F}_n^* \cdot \boldsymbol{\epsilon}(\mathbf{r}) \cdot \mathbf{F}_n = 1$ |
| Inhomogeneous, lossless, weakly dispersive, anisotropic | $\sum_n \sqrt{\frac{\hbar}{2\epsilon_0\omega_n}} \left(\mathbf{F}_n(\mathbf{r}) e^{-i\omega_n t} a_n + \mathbf{F}_n^*(\mathbf{r}) e^{i\omega_n t} a_n^\dagger \right)$ | $\nabla \times \nabla \times \mathbf{F}_n(\mathbf{r}) = \boldsymbol{\epsilon}(\mathbf{r}, \omega_n) \frac{\omega_n^2}{c^2} \mathbf{F}_n(\mathbf{r});$ $\frac{1}{2\omega_n} \int d^3r \mathbf{F}_n^*(\mathbf{r}) \cdot \frac{d(\omega^2 \boldsymbol{\epsilon}(\mathbf{r}, \omega))}{d\omega} \Big _{\omega_n} \cdot \mathbf{F}_n(\mathbf{r}) = 1$ |
| Inhomogeneous, lossy, dispersive, isotropic, local | $\sqrt{\frac{\hbar}{\pi\epsilon_0}} \int_0^\infty d\omega \int d^3r' \frac{\omega}{c^2} \sqrt{\text{Im } \boldsymbol{\epsilon}(\mathbf{r}', \omega)}$ $(\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \mathbf{f}(\mathbf{r}', \omega) e^{-i\omega t} + \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) \mathbf{f}^\dagger(\mathbf{r}', \omega) e^{i\omega t})$ | $\left(\nabla \times \nabla \times - \boldsymbol{\epsilon}(\mathbf{r}, \omega) \frac{\omega^2}{c^2} \right) \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') I$ |
| Inhomogeneous, lossy, dispersive, anisotropic, non-local | $\sqrt{\frac{\hbar}{\pi\epsilon_0}} \int_0^\infty d\omega \int d^3r' \frac{\omega}{c^2} \sqrt{\text{Im } \boldsymbol{\epsilon}(\mathbf{r}, \mathbf{r}', \omega)}$ $(\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \mathbf{f}(\mathbf{r}', \omega) e^{-i\omega t} + \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) \mathbf{f}^\dagger(\mathbf{r}', \omega) e^{i\omega t})$ | $\left(\nabla \times \nabla \times - \int d^3r' \boldsymbol{\epsilon}(\mathbf{r}, \mathbf{r}', \omega) \frac{\omega^2}{c^2} \right) \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') I$ |

Additional details on the general framework for nanophotonic scintillation & experiments

Roques-Carmes*, **Rivera*** *et al.* *A general framework for scintillation in nanophotonics.* *arXiv: 2110.11492.*

A general framework for describing nanophotonic scintillators

- Time-scale separation leads to an approximate non-equilibrium distribution
- Spectral & angular density of emitted light in scintillation

$$\frac{dP^{(i)}}{d\omega d\Omega} = \frac{\omega^2}{8\pi^2 \epsilon_0 c^3} \int d\mathbf{r}' \frac{E_j^{*(i)}(\mathbf{r}', \mathbf{r}, \omega)}{|\mathbf{E}_{\text{inc}}^{(i)}(\mathbf{r}', \mathbf{r}, \omega)|} \frac{E_k^{(i)}(\mathbf{r}', \mathbf{r}, \omega)}{|\mathbf{E}_{\text{inc}}^{(i)}(\mathbf{r}', \mathbf{r}, \omega)|} S_{jk}(\mathbf{r}', \omega)$$

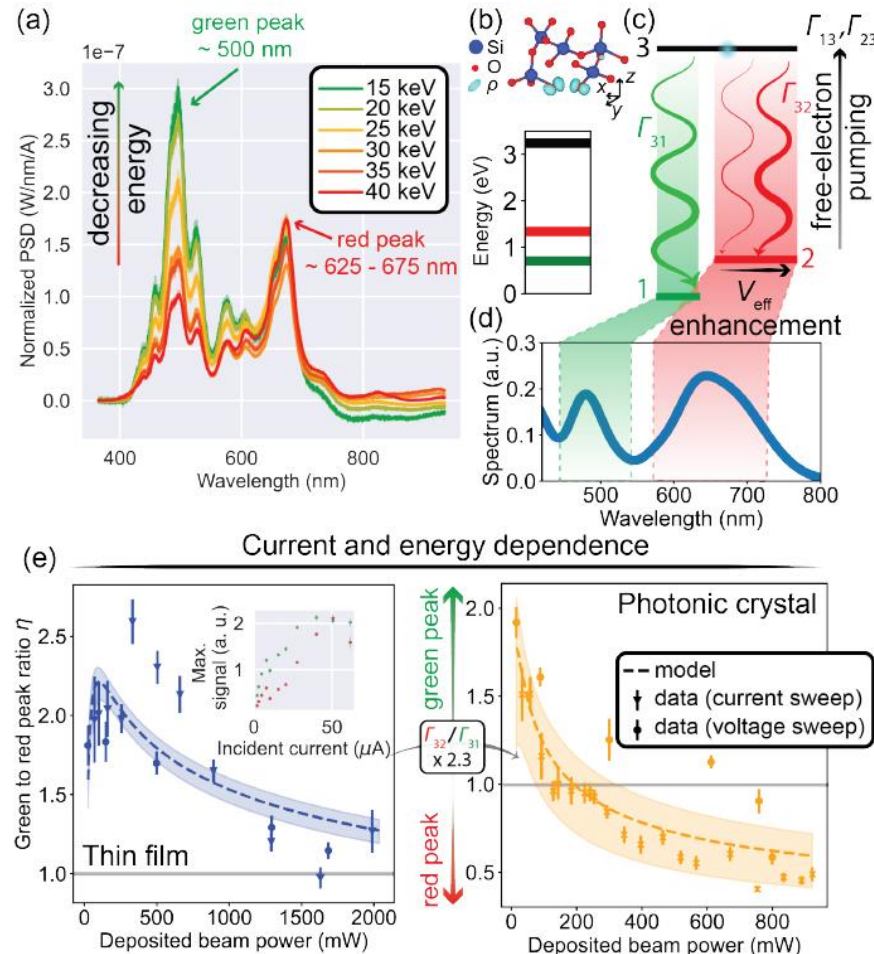
[specified by Maxwell solutions]

$$S_{jk}(\mathbf{r}_1, \mathbf{r}_2, \omega) \sim \langle J_j^-(\mathbf{r}_1, \omega) J_k^+(\mathbf{r}_2, \omega) \rangle$$

[specified by microscopics: energies, matrix elements, occupations]

- Some similarity to description of thermal radiation (but non-equilibrium electrons require more)

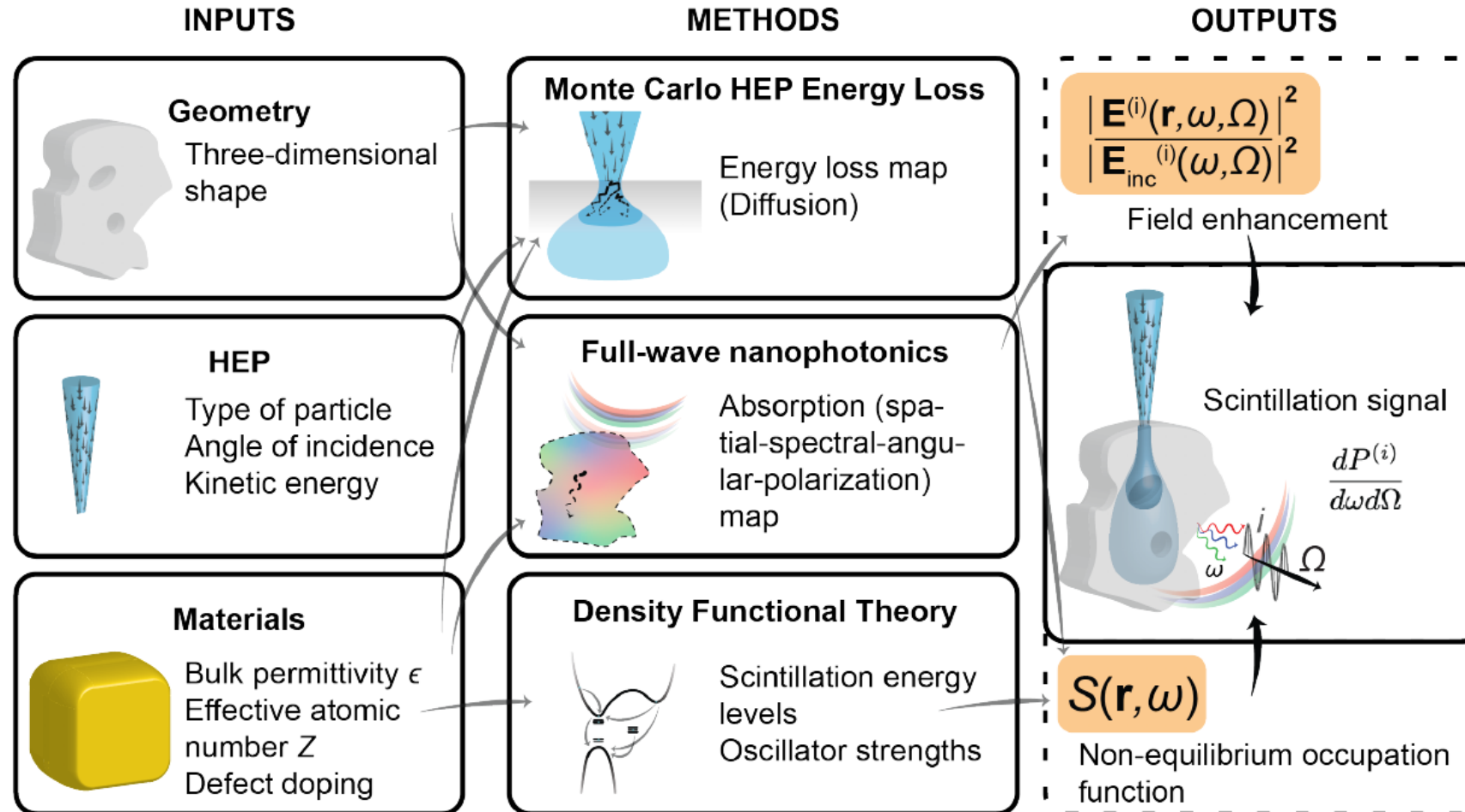
Out-of-equilibrium physics of defects controlled by nanophotonics



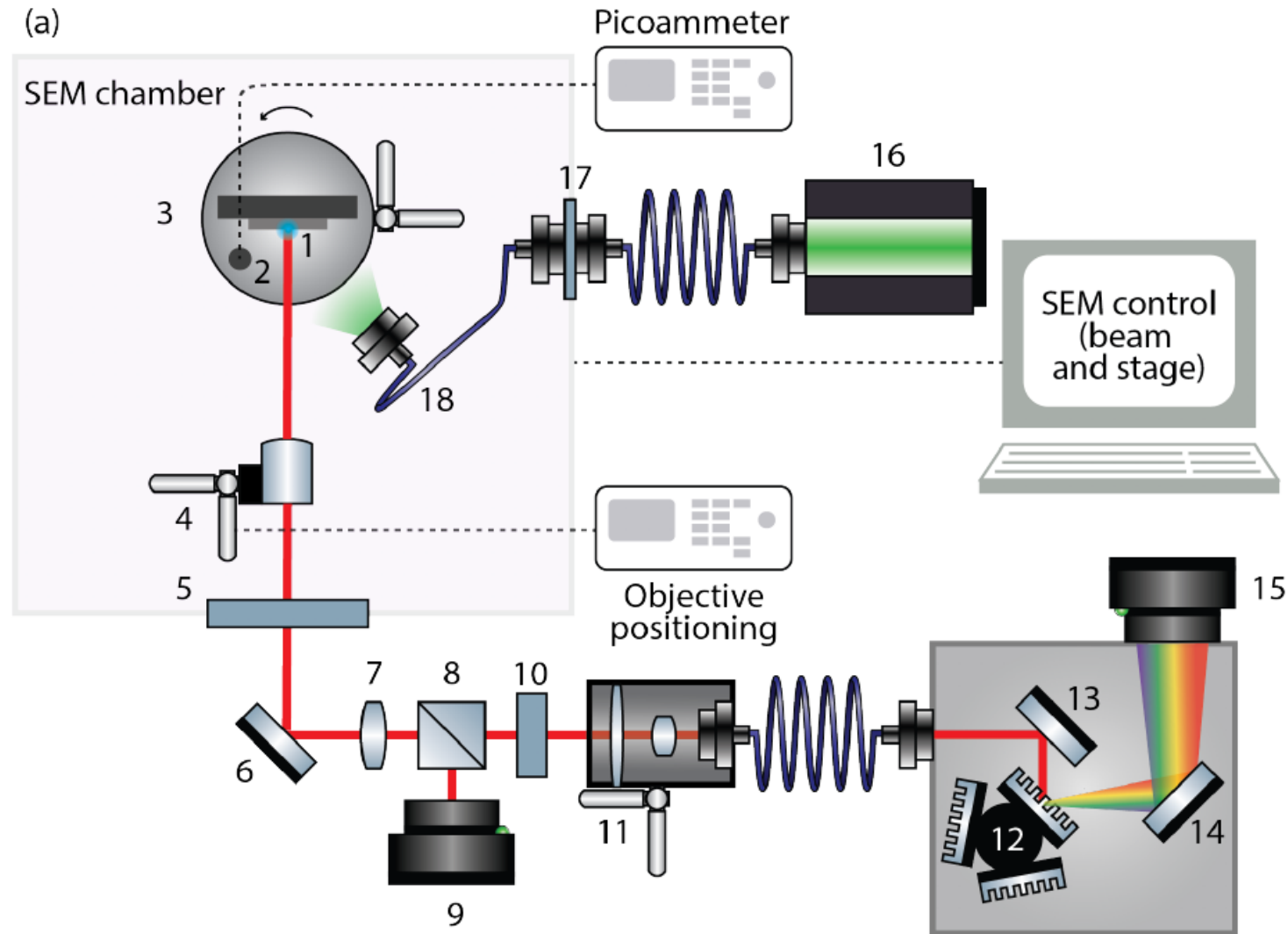
- Beyond predicting “nanophotonic” features the framework also, accounts for features requiring microscopics+nanophotonics
 - Nonlinear dependence of emission on pump + “cross-over” from green to red
 - 3-level dynamics of self-trapped holes

$$\begin{cases} \frac{dp_1}{dt} = -\Gamma_{13} p_1 (1 - p_3) + \Gamma_{31} p_3 (1 - p_1) \\ \frac{dp_2}{dt} = -\Gamma_{23} p_2 (1 - p_3) + \Gamma_{32} p_3 (1 - p_2) \\ \frac{dp_3}{dt} = \Gamma_{13} p_1 (1 - p_3) - \Gamma_{31} p_3 (1 - p_1) \\ \quad + \Gamma_{23} p_2 (1 - p_3) - \Gamma_{32} p_3 (1 - p_2) \end{cases}$$

The end-to-end numerical pipeline for nanophotonic scintillation, summarized



Electron-beam scintillation setup

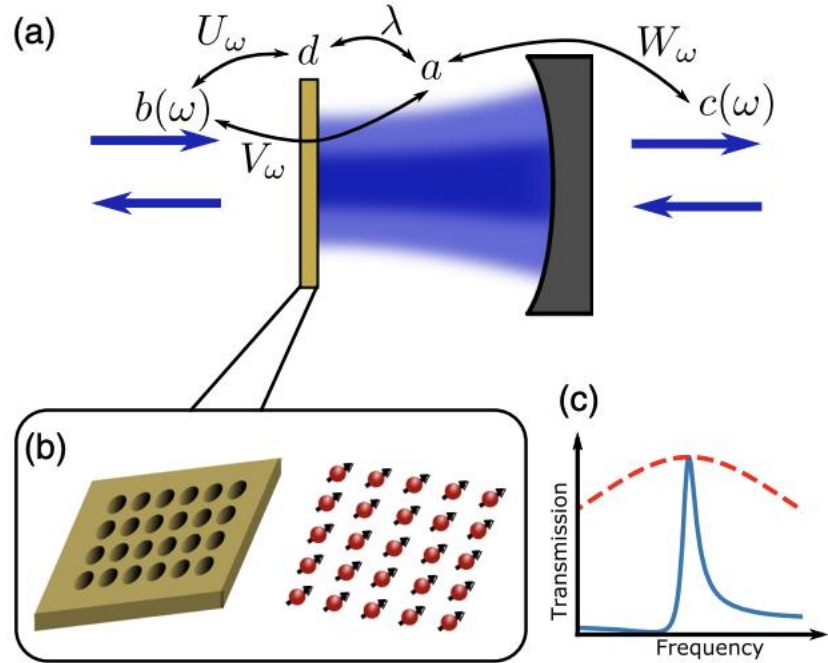


Theory of nonlinear lasers and transient Fock state generation

Rivera et al. Macroscopic condensation of photon noise in sharply nonlinear dissipative systems. In preparation.

A “microscopic model” of nonlinear loss

Inspiration: Cernotik *et al.* *PRL* (2019).



- Could we find the posited dynamics resulting from the exact description of a system?
 - Need a quantum optical model (Lindblad theory) of a frequency-dependent reflector.
- Compared to Cernotik *et al.*: we consider the case of an *anharmonic* oscillator undergoing dissipation, as would be from inserting a nonlinear medium.

$$\begin{aligned}
 H/\hbar = & H_a + \omega_d d^\dagger d + (\lambda a d^\dagger + \lambda^* a^\dagger d) + \sum_k \omega_k b_k^\dagger b_k \\
 & + \sum_k (g_k a b_k^\dagger + g_k^* a^\dagger b_k) + \sum_k (v_k d b_k^\dagger + v_k^* d^\dagger b_k).
 \end{aligned}$$

Rivera *et al.* *In preparation.*

- Model of Fano resonance (asymmetric scattering lineshape; great recent interest [see Zhen *et al.* *Nat. Rev. Mat.* (2016)])

Lindbladian for an anharmonic oscillator coupled to a frequency-dependent loss

- When two systems (a, d) are coupled to the same reservoir, the jump/transition operator in the Lindbladian describing loss couples

$$\dot{\rho}_{ad} = -i[H_{ad}/\hbar, \rho_{ad}] + (X^\dagger X \rho_{ad} + \rho_{ad} X^\dagger X - 2X \rho_{ad} X^\dagger).$$

$$X \equiv \sqrt{\kappa}a + \sqrt{\gamma}d.$$

- In the adiabatic limit, where d (mirror mode) decays much faster than the round-trip time, we adiabatically eliminate d from Liouvillian
 - This enables us to find an EOM for a
 - Also verified numerically (“exactly”)

Rivera et al. In preparation.

$$\begin{aligned} \dot{\rho} = & -\kappa(a^\dagger a \rho + \rho a^\dagger a - 2a \rho a^\dagger) \\ & + \sum_{n=0}^{\infty} \frac{n G_+ G_-}{i(\omega_d - \omega_{n,n-1}) + \gamma} T_{n,n} \rho \\ & + \sum_{n=0}^{\infty} \frac{n (G_+ G_-)^*}{-i(\omega_d - \omega_{n,n-1}) + \gamma} \rho T_{n,n} \\ & + \sum_{m,n=0}^{\infty} \frac{\sqrt{m(n+1)} (G_+ G_-)^*}{-i(\omega_d - \omega_{n+1,n}) + \gamma} T_{m-1,m} \rho T_{n+1,n} \\ & + \sum_{m,n=0}^{\infty} \frac{\sqrt{m(n+1)} (G_+ G_-)}{i(\omega_d - \omega_{m,m-1}) + \gamma} T_{m-1,m} \rho T_{n+1,n} \end{aligned}$$



$$\begin{aligned} \dot{\rho}_{n,n} = & -n \left(\kappa - 2\text{Re} \left[\frac{G_+ G_-}{i(\omega_d - \omega_{n,n-1}) + \gamma/2} \right] \right) \rho_{n,n} + \\ & (n+1) \left(\kappa - 2\text{Re} \left[\frac{G_+ G_-}{i(\omega_d - \omega_{n+1,n}) + \gamma/2} \right] \right) \rho_{n+1,n+1}, \end{aligned}$$

Description of the nonlinear loss in terms of a number-dependent Langevin forces

- We can associate a Langevin equation with the density matrix equation

$$\dot{T}_{n,n} = -L_n T_{n,n} + L_{n+1} T_{n+1,n+1} + F_{n,n}$$

- Which, from the Einstein relation: $2\langle D_{\mu\nu} \rangle = \frac{d}{dt} \langle A_\mu A_\nu \rangle - \langle A_\mu D_\nu \rangle - \langle D_\mu A_\nu \rangle$ implies

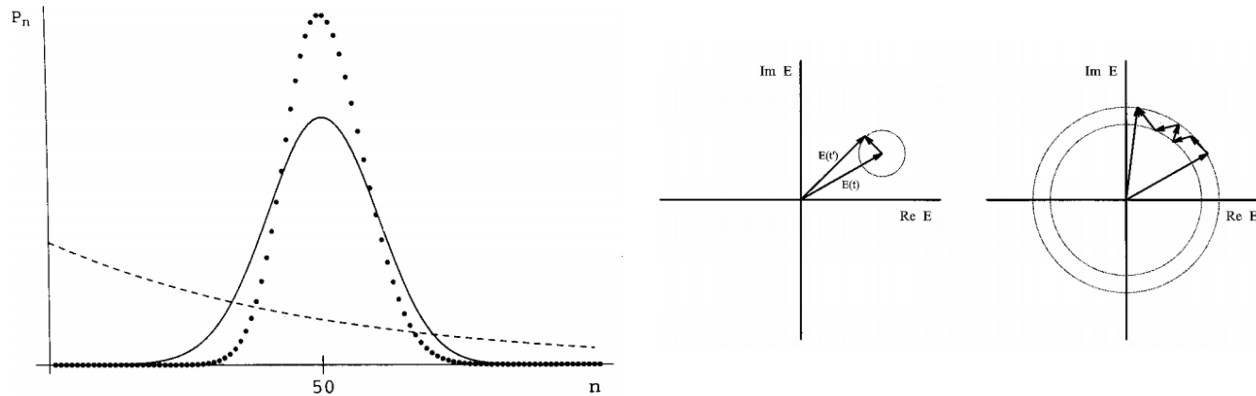
$$2\langle D_{n,n} \rangle = \langle L(n) \rangle = \langle n\kappa(n) \rangle$$

- This is then converted to a Langevin equation for the photon number

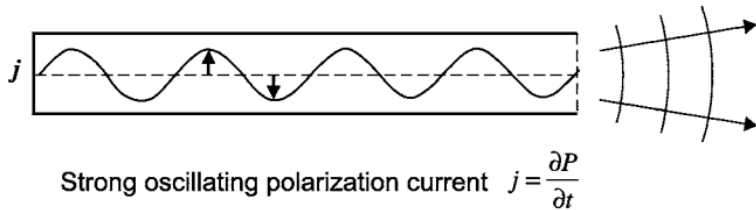
$$\dot{n} = -\kappa(n)n + F_n \quad \text{with} \quad \begin{aligned} \langle F_n(t) \rangle &= 0 \\ \langle F_n(t)F_n(t') \rangle &= \langle L(n) \rangle \delta(t - t'). \end{aligned}$$

- Advantage of Langevin: easy to get moments, noise spectrum (corresponds to a common experimental observable), especially with important classes of gain (solid-state & semiconductor lasers)

Relation to “textbook” quantum laser theory, e.g., of Lamb & Scully



Glauber, *Nobel Lecture* (2005)



- “Textbook result”
 - “De-phased” coherent states (well-beyond threshold)
- Here, saturation comes from photon NL rather than gain
 - Here, we develop a new quantum theory of these “nonlinear lasers”
 - Covers many more cases than presented here

Relation to “textbook” quantum laser theory, e.g., of Lamb & Scully

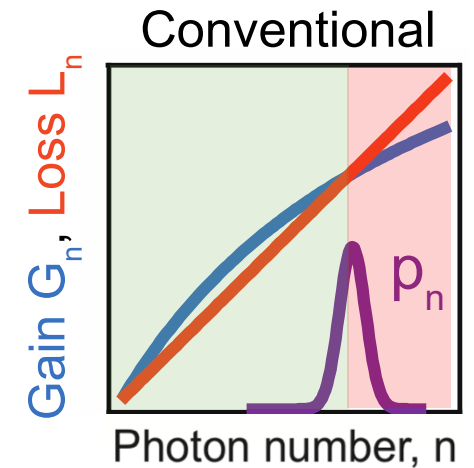
- Dynamics of the resonator photon probability distribution p_n can be shown to follow a rate equation

$$\dot{p}_n = G_n p_{n-1} - (L_n + G_{n+1}) p_n + L_{n+1} p_{n+1}.$$

- Textbook case of the quantum optics of a laser:

- $R_{\text{emit}}(n - 1 \rightarrow n) = \frac{An}{1+n/n_s} = G_n,$
- $R_{\text{loss}}(n \rightarrow n - 1) = \kappa n = L_n$

- Key to the “coherent” state: ”negative feedback” from atomic saturation (as intensity increases, gain decreases)



$$\Delta n = \left(1 - G_{\langle n \rangle + 1} / L_{\langle n \rangle + 1}\right)^{-1/2}$$

Infinite-order nonlinearities and Fock-state lasing in the deep-strong coupling regime of QED

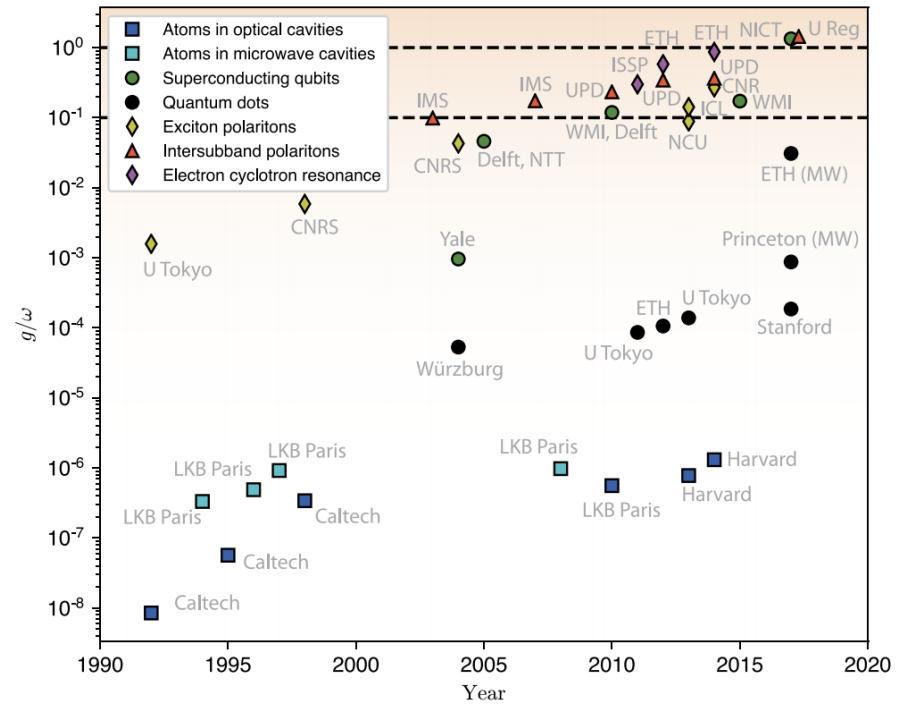
Rivera et al. Infinite-order nonlinearities and Fock-state lasing in the deep-strong coupling regime of QED.

These effects may also be realized using exotic new excitations in cavity QED

TLS coupled to a cavity

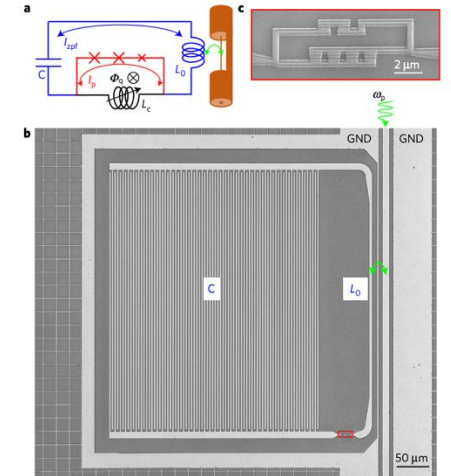
$$H_R = \left(\frac{\hbar\omega_0}{2}\sigma_z + \frac{\hbar\lambda}{2}\sigma_x \right) + \hbar\omega a^\dagger a + \hbar\tilde{g}\sigma_x(a + a^\dagger)$$

Deep-strong coupling (DSC) regime (coupling \gg bare energies)

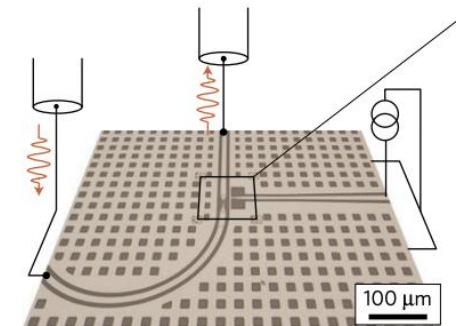


Forn-Díaz, *et al. Rev. Mod. Phys.* (2019).

Devoret, *et al. Ann. Phys.* (2007).

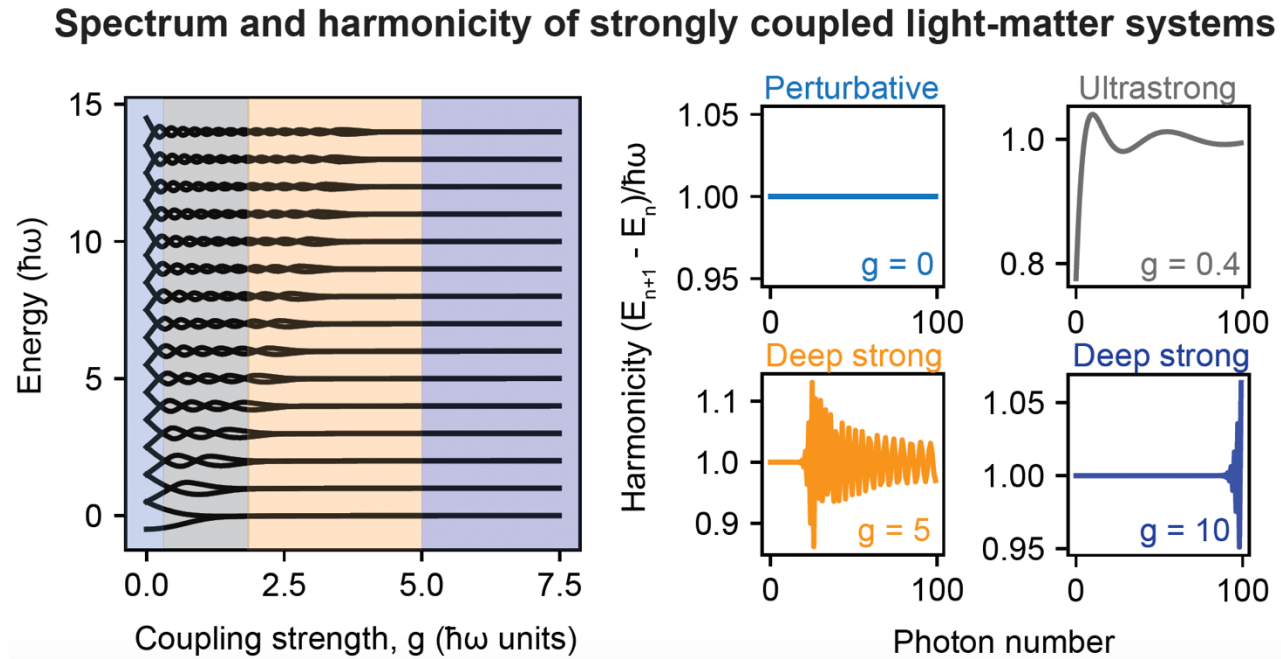


Forn-Díaz, *et al. Nat. Phys.* (2017).



Yoshihara, *et al. Nat. Phys.* (2017).

An extremely high-order nonlinearity arising from a TLS coupling to a cavity

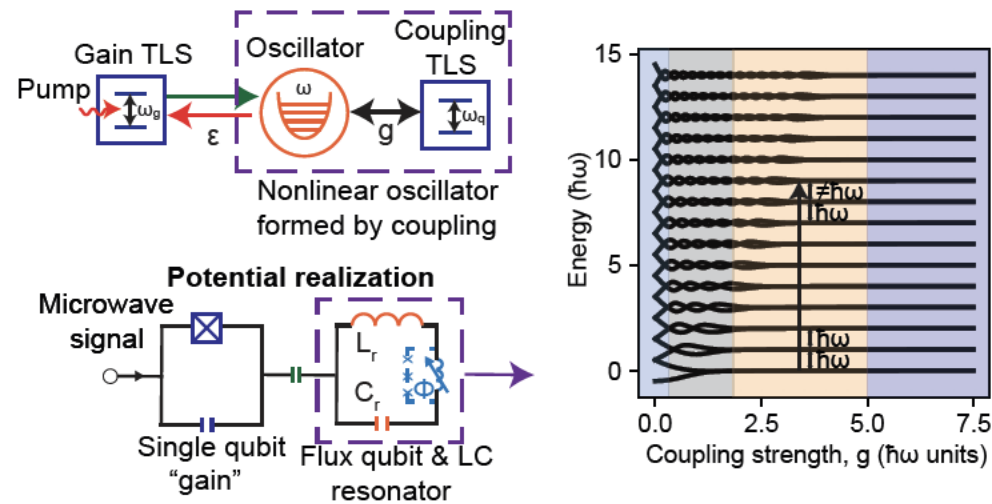


- The eigenstates are an effective photon (DSC photon) with a pseudo-spin

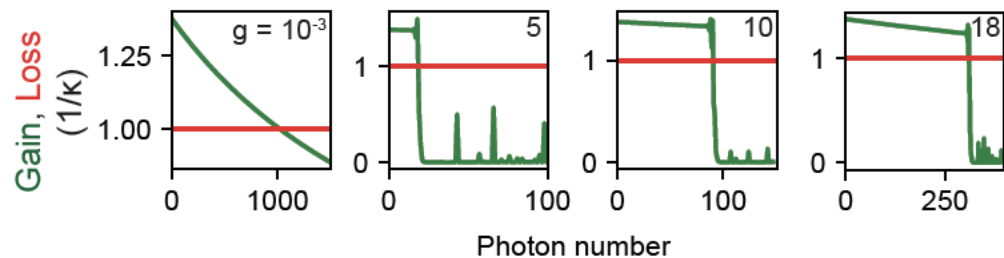
$$E_{n\sigma} = \hbar\omega \left(n + \frac{1}{2} \sigma e^{-2|g|^2} L_n(4|g|^2) \right) \quad |n, \sigma\rangle = \frac{1}{\sqrt{2}} (D^\dagger | + x, n\rangle + \sigma D | - x, n\rangle)$$

These form the basis for a Fock maser, for example, with sharp NL gain

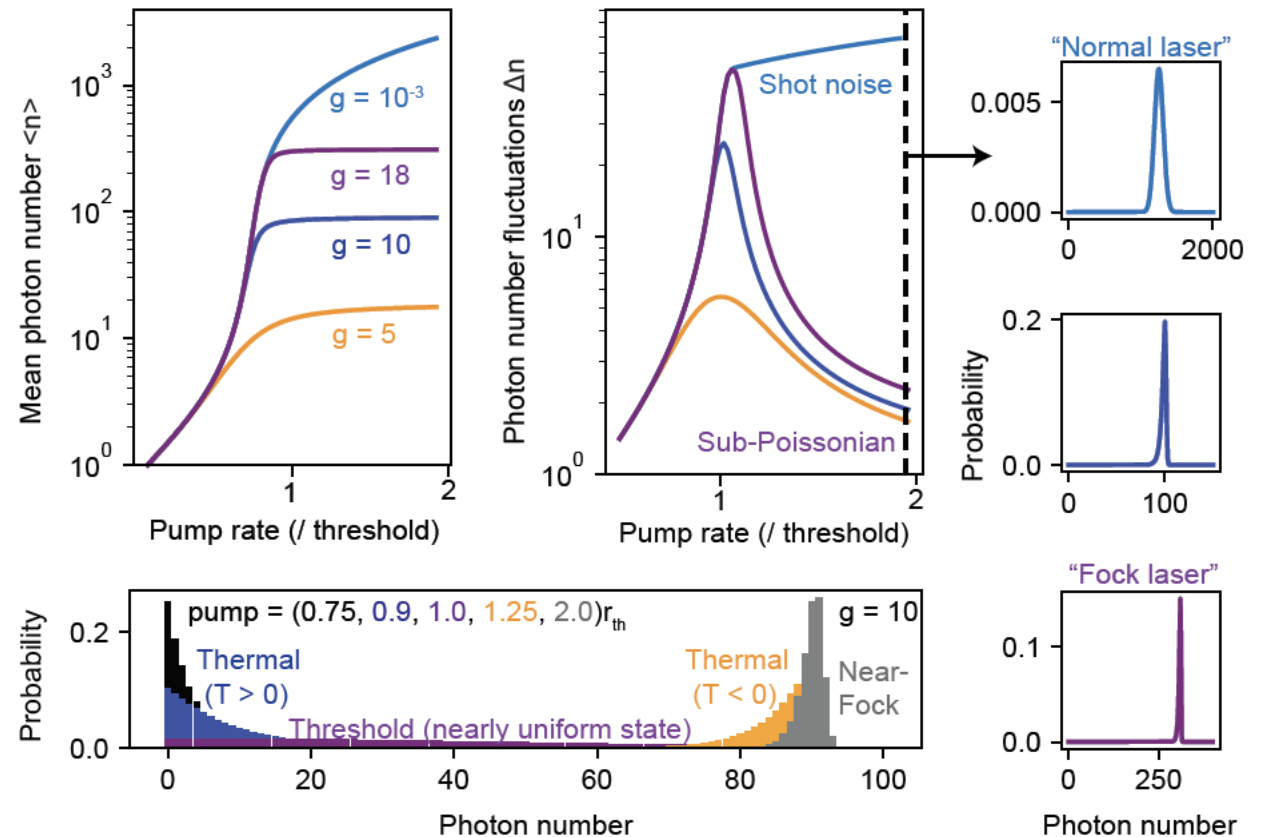
(a) Example realization of Fock lasing based on sharp gain



(b) Nonlinear gain in the deep-strong coupling laser



(c) Laser output and quantum statistics



Stimulated emission of DSC photons

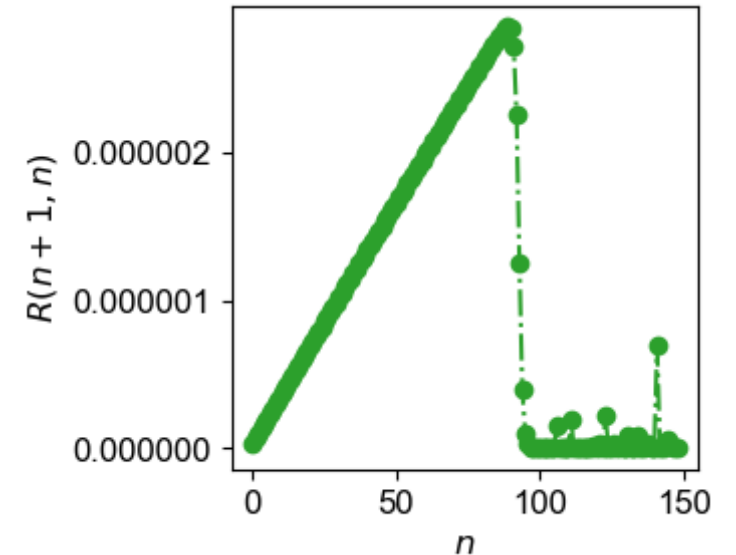
- Consider an ancillary TLS coupled to the field of the DSC photon through the JC interaction: $\epsilon\sigma_{x,\text{em}}(a + a^\dagger)$

- The stimulated emission rate is found to be:

$$R_{(n+1)\sigma',n\sigma}^{(\epsilon)} \approx \frac{2r_a\epsilon^2(n+1)}{\Gamma^2 + 4\epsilon^2(n+1) + \Delta_{n+1}^2} \delta_{\sigma,\sigma'}$$

- Notice the similarity to textbook laser physics:

$$\chi = -\frac{i\chi_0\gamma[\gamma - i(\omega-\nu)]}{\gamma^2(1 + I/I_s) + (\omega-\nu)^2}.$$



Gain dynamics of a laser based on emission into DSC photons

- For the photon probabilities $p(n)$ of having n photons:

$$\dot{p}_n = G_n p_{n-1} - (L_n + G_{n+1}) p_n + L_{n+1} p_{n+1}$$

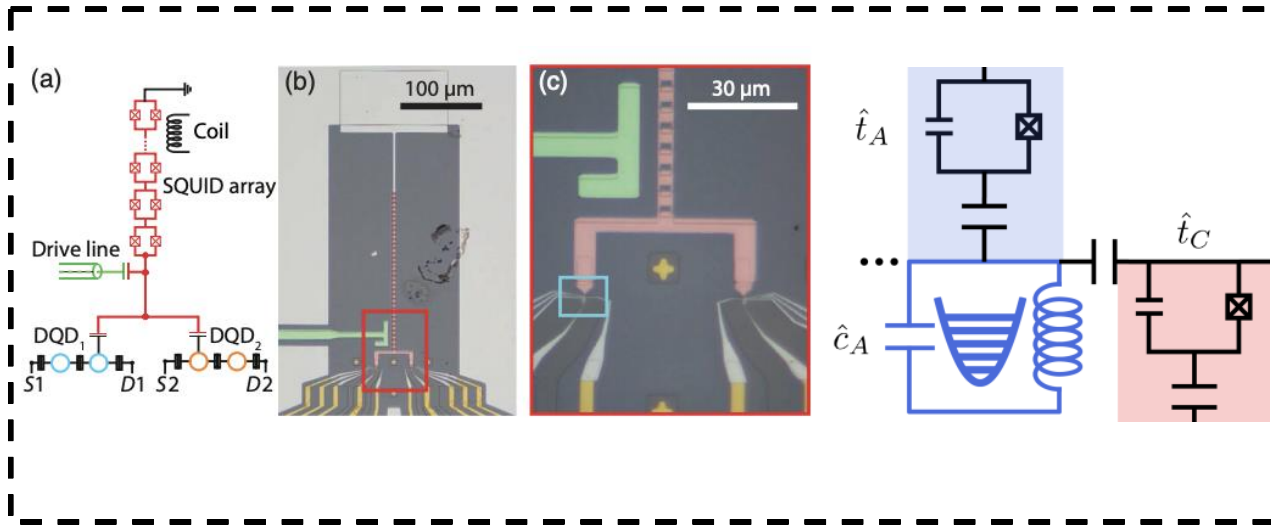
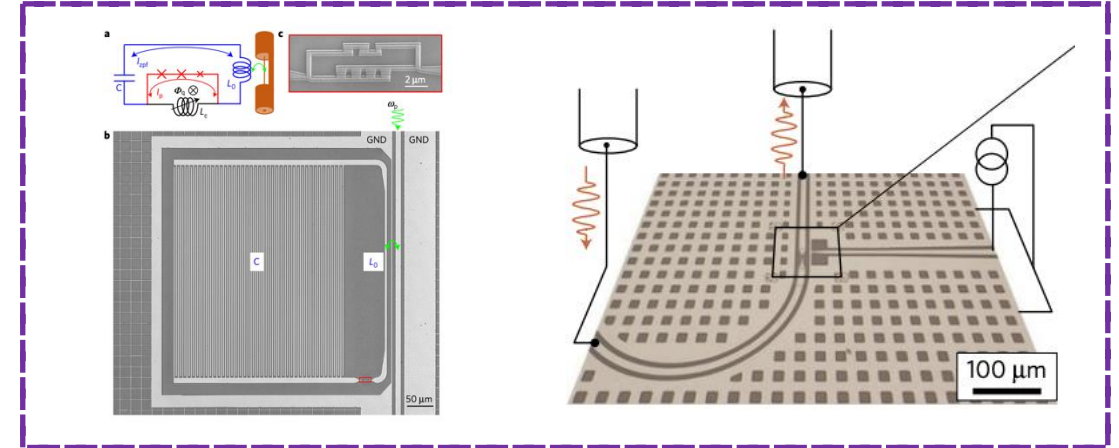
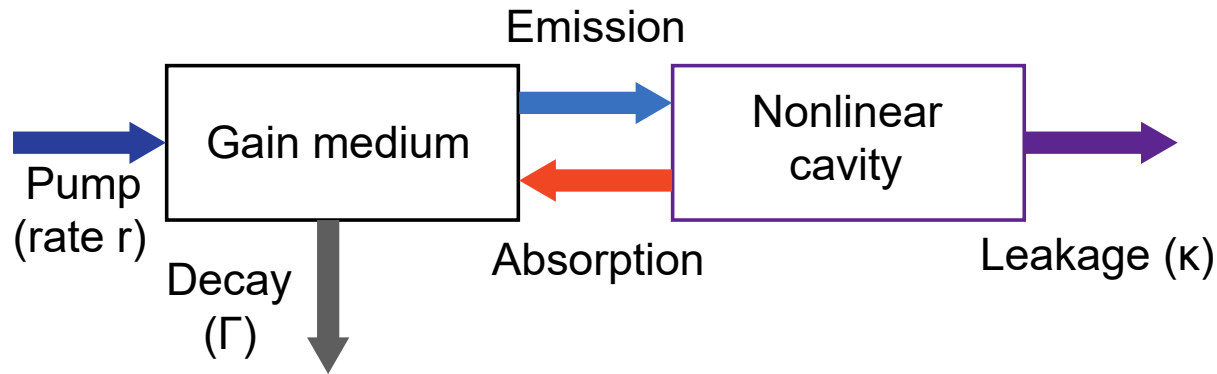
- Which, for our problem, reads

$$\dot{\rho}_{nn} = \frac{2rn\epsilon^2}{\Gamma^2 + F(n)} \rho_{n-1,n-1} - \left(\frac{2r(n+1)\epsilon^2}{\Gamma^2 + F(n+1)} + \kappa n \right) \rho_{nn} + \kappa(n+1) \rho_{n+1,n+1}$$

- where the nonlinearity $F(n) = 4n\epsilon^2 + \frac{1}{4}\omega^2 e^{-4g^2} (L_n(4g^2) - L_{n-1}(4g^2))^2$

- The steady-state solution is: $\rho_n = \frac{1}{Z} \prod_{m=1}^n \frac{2r\epsilon^2/\kappa}{\Gamma^2 + F(m)}$

Paths to experimental realization

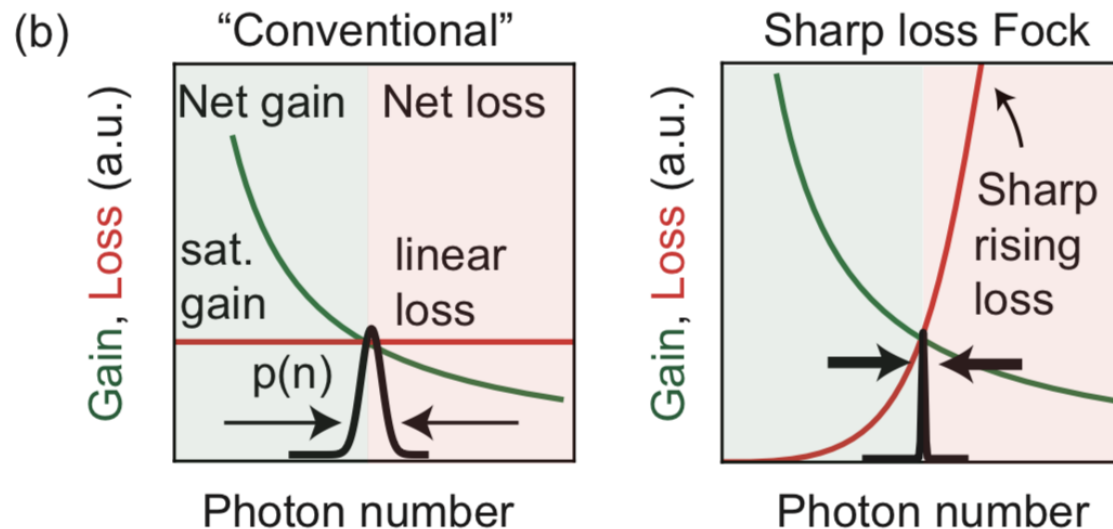
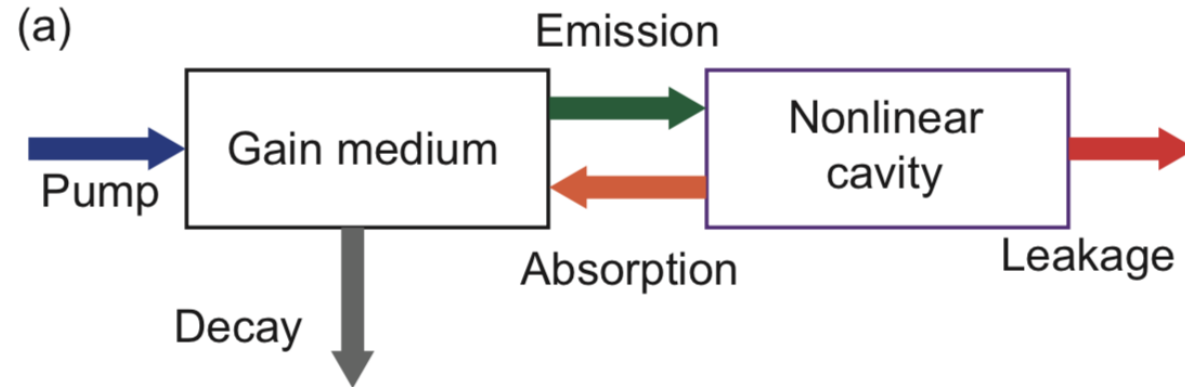


- A key realization from previous slide: “gain” can be provided by a single emitter,
 - a repeatedly pumped qubit
 - or multiple emitters, such as microwave-active molecules

Condensation of optical noise in the macroscopic regime

Rivera et al. *Macroscopic condensation of photon noise in sharply nonlinear dissipative systems. In preparation.*

Noise properties of lasers with nonlinear dissipation



- Presence of saturable gain adds terms to master eqn. (Lamb, Scully)

$$\dot{p}_n = G_n p_{n-1} - (L_n + G_{n+1}) p_n + L_{n+1} p_{n+1}.$$

- Admits steady-state solution:

$$p_n = \frac{1}{Z} \prod_{m=1}^n \frac{G_m}{L_m}$$

- Noise given by

$$\Delta n = \left(1 - G_{\langle n \rangle + 1} / L_{\langle n \rangle + 1}\right)^{-1/2}$$

Condensation of optical noise in the macroscopic regime?

- Master eq. for probabilities corresponds to a Langevin equation for the photon number $\dot{n} = -\kappa(n)n + F_n$. with F_n an operator-valued, Langevin force with statistical properties dictated by the Einstein relation (FDT)

$$\langle F_n(t) \rangle = 0$$

$$\langle F_n(t)F_n(t') \rangle = \langle L(n) \rangle \delta(t - t')$$

- Combining this with a description of the inversion a la Lax, we have:

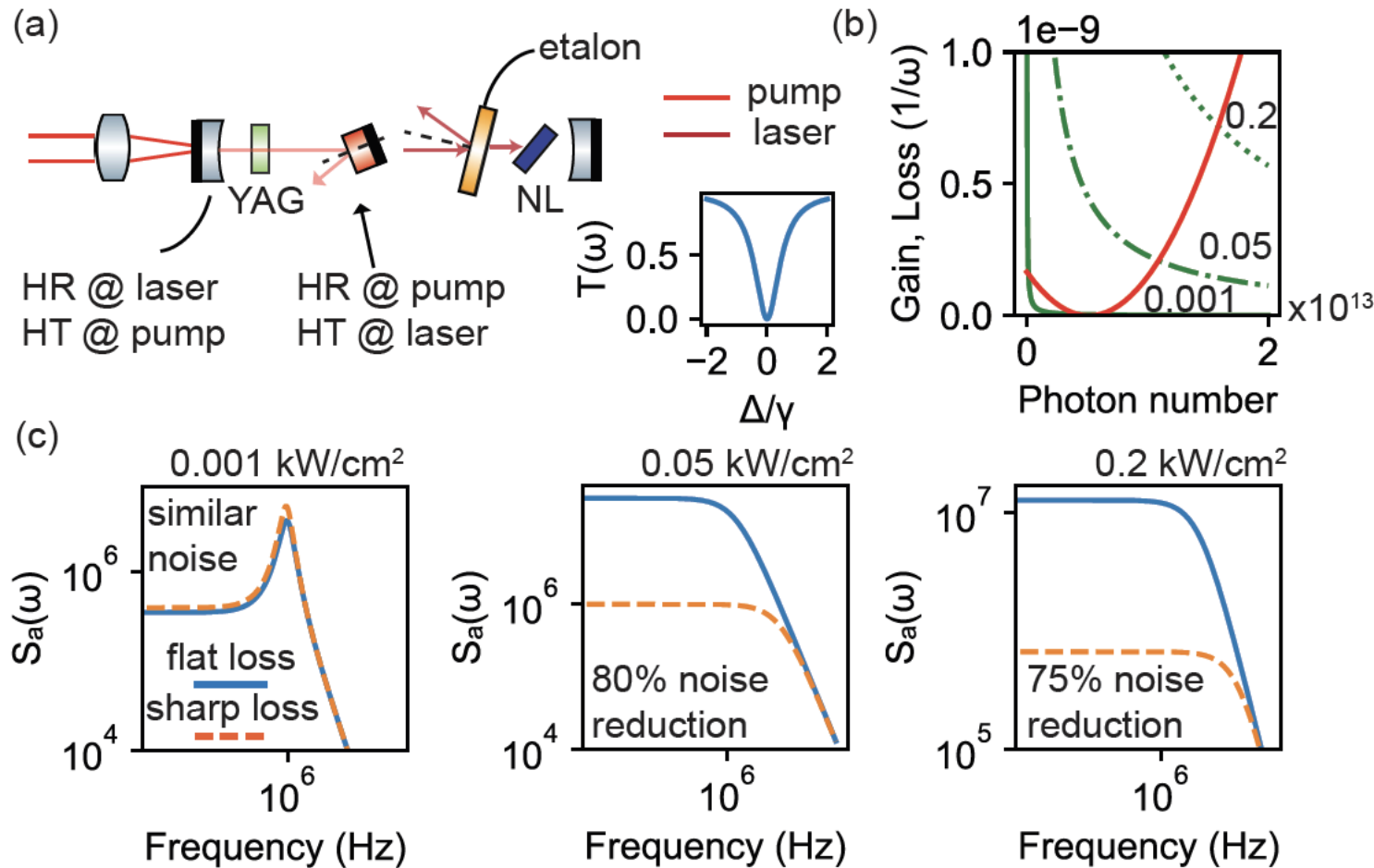
$$\dot{n} = (R_{\text{sp}}S - \kappa(n))n + F_n$$

$$\dot{S} = \Lambda - (\gamma_{\parallel} + R_{\text{sp}}n)S + F_S$$



$$\begin{pmatrix} \delta \dot{S} \\ \delta \dot{n} \end{pmatrix} = \begin{pmatrix} -(\gamma_{\parallel} + R_{\text{sp}}\langle n \rangle) & -R_{\text{sp}}\langle S \rangle \\ R_{\text{sp}}\langle n \rangle & -\kappa'(\langle n \rangle)\langle n \rangle \end{pmatrix} \begin{pmatrix} \delta S \\ \delta n \end{pmatrix} + \begin{pmatrix} F_S \\ F_n \end{pmatrix}$$

Condensation of optical noise in the macroscopic regime?



Spectrum of number fluctuations

$$S_a(\omega) = \frac{1}{\pi} \langle \delta n^\dagger(\omega) \delta n(\omega) \rangle$$

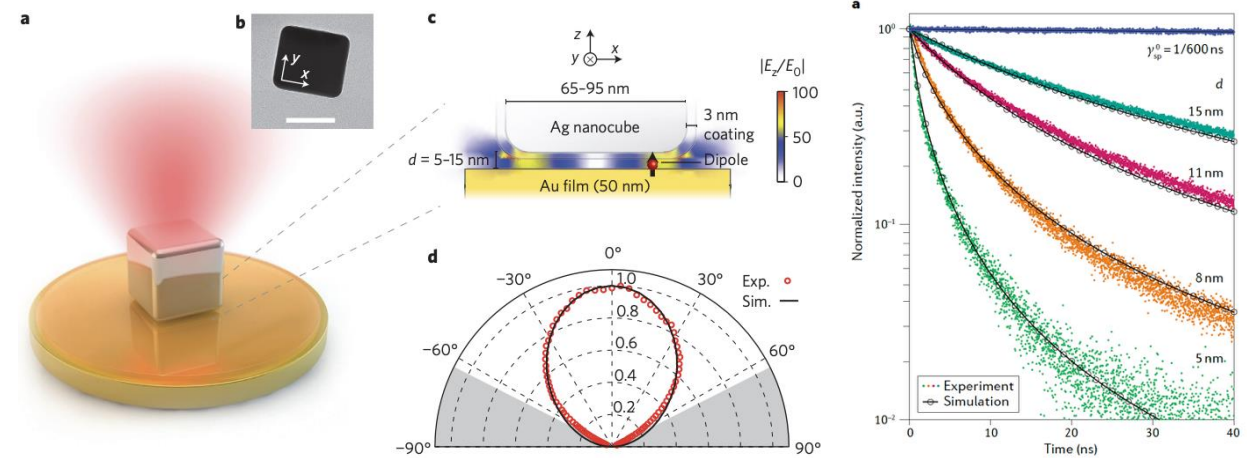
related to overall noise via

$$(\Delta n)^2 = \int_0^\infty d\omega S_a(\omega)$$

Note: incomplete condensation because the NL here is weak, but we still get a fairly strong effect due to inclusion of a very sharp freq.-dependent element.

Confining light on wavelength scales to control light-matter interaction

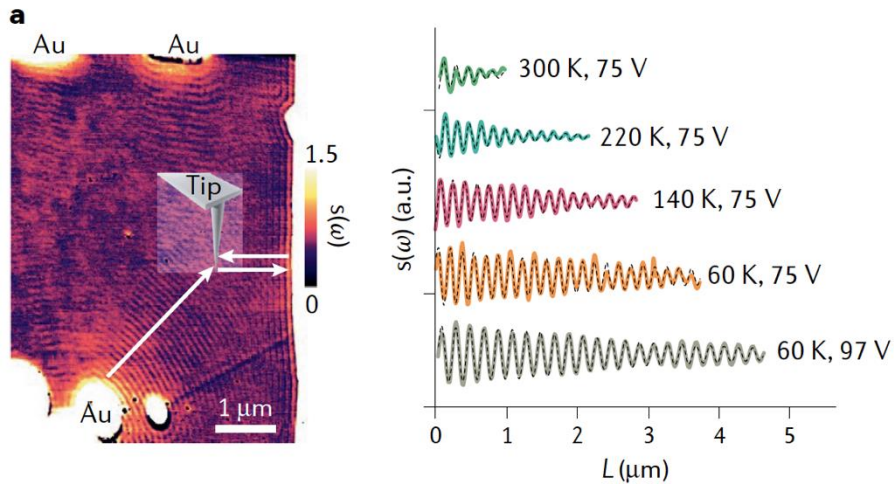
- Many of the applications above: light is a *plane wave*, with energy delocalized over a volume $\gg \lambda^3$.
- By controlling light with polarizable media with spatial variations on the scale of λ (*nanophotonics*), we can enhance the interactions.
- Very representative: Purcell effect
- **Still, the interactions are very weak, and are largely lowest-order (e.g., in coupling strength, atomic size, field strength)**



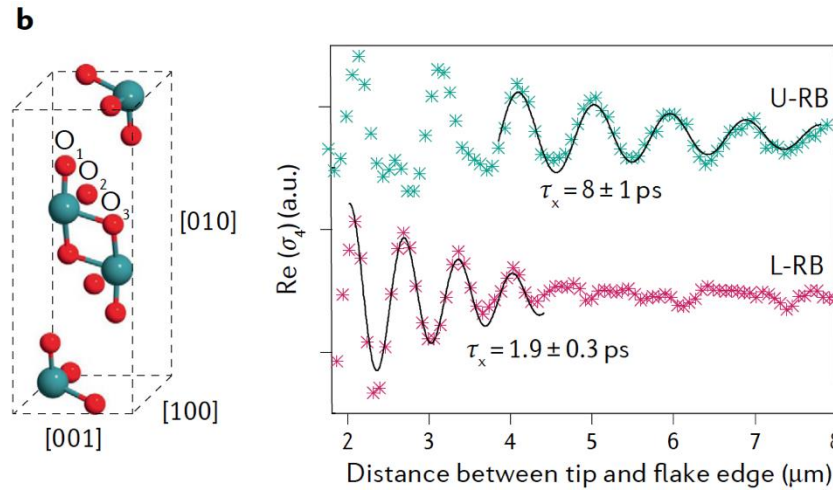
Akselrod *et al.* *Nature Photonics* (2014).

$$\Gamma_{fi} = \frac{3}{4\pi^2} \frac{Q}{(V/\lambda_0^3)} |\hat{\mathbf{d}}_{fi} \cdot \mathbf{u}(\mathbf{r})|^2 \Gamma_0,$$

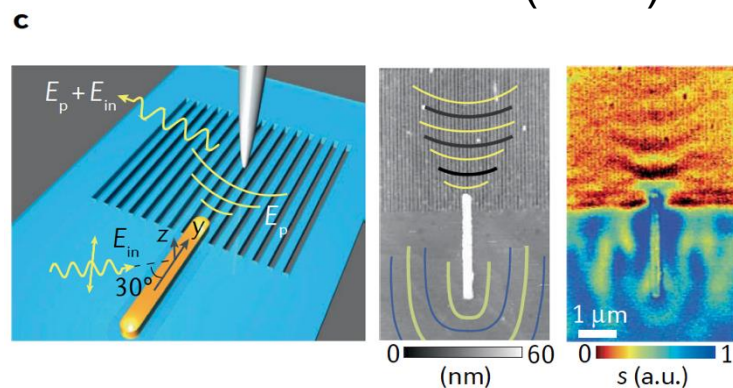
Collective excitations as generalized photons, or photonic quasiparticles



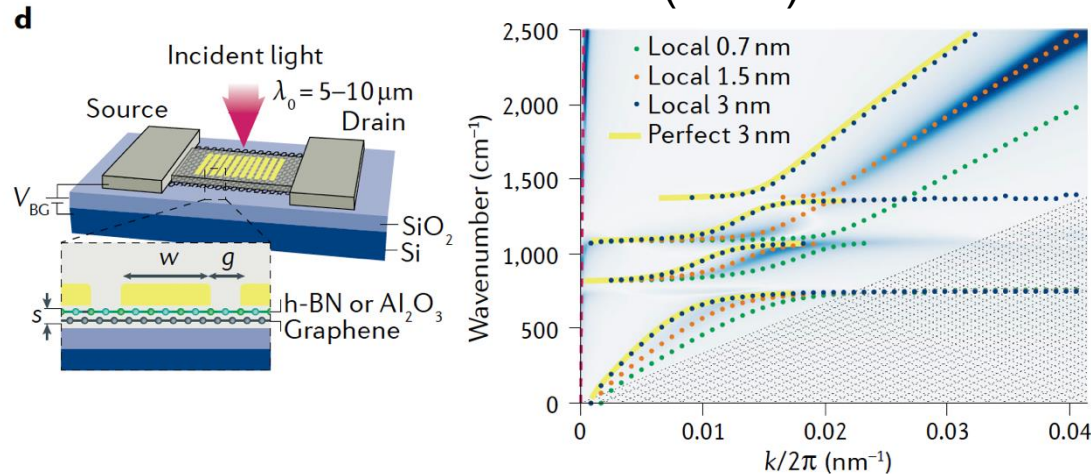
Ni et al. Nature (2018).



Ma et al. Nature (2018).



Li et al. Science (2018).



Iranzo et al. Science (2018).

Can be used in many of the same ways as photons, but very different linear properties:

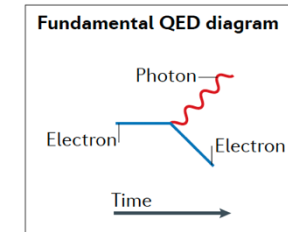
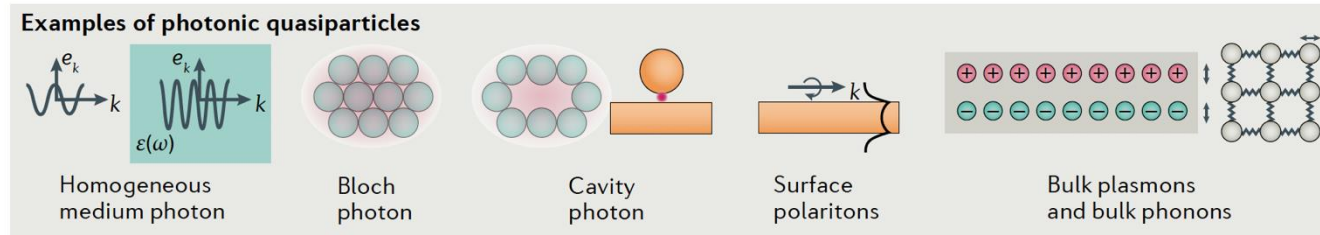
- *Wavelength* – >100x smaller
- *Polarization* – often circular
- *DOS* – very high
- *Mode volume* – very small

Outline

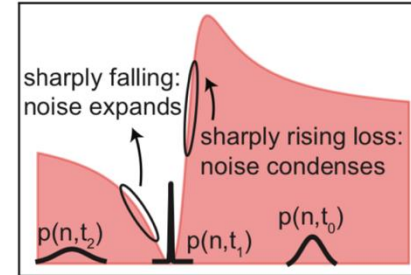
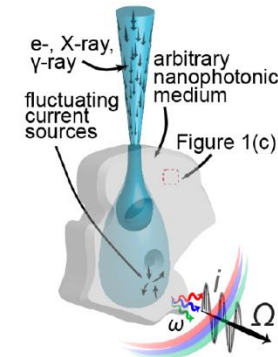
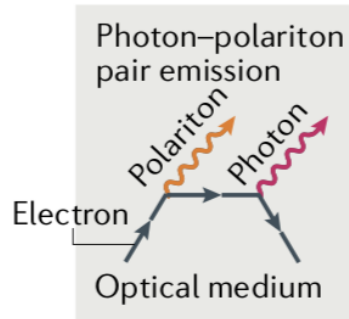
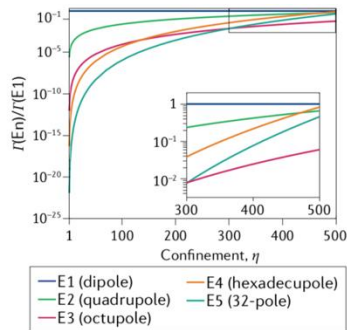
1. Intro to light-matter interactions and photonic quasiparticles
2. Systematic study of elementary processes of light-matter interactions with photonic quasiparticles
3. An application of enhancing light-matter interactions – novel high-energy particle detectors based on scintillators
4. Photonic quasiparticle nonlinearities, new nonlinear dissipative phenomena, and large optical Fock state generation

Putting it all together

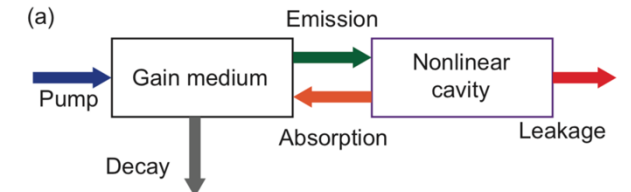
Light-matter interactions with photonic quasiparticles



Interactions arising from linear properties



& nonlinear properties

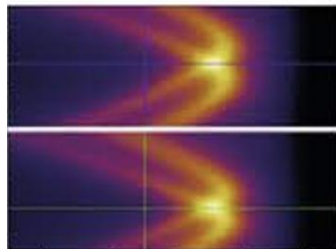


Some potential applications of these interactions

Detectors



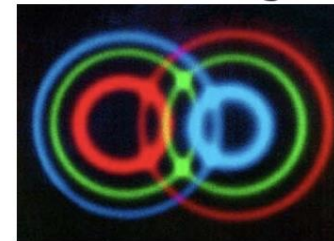
Spectroscopy



Particle detection



Quantum light



Lasers

