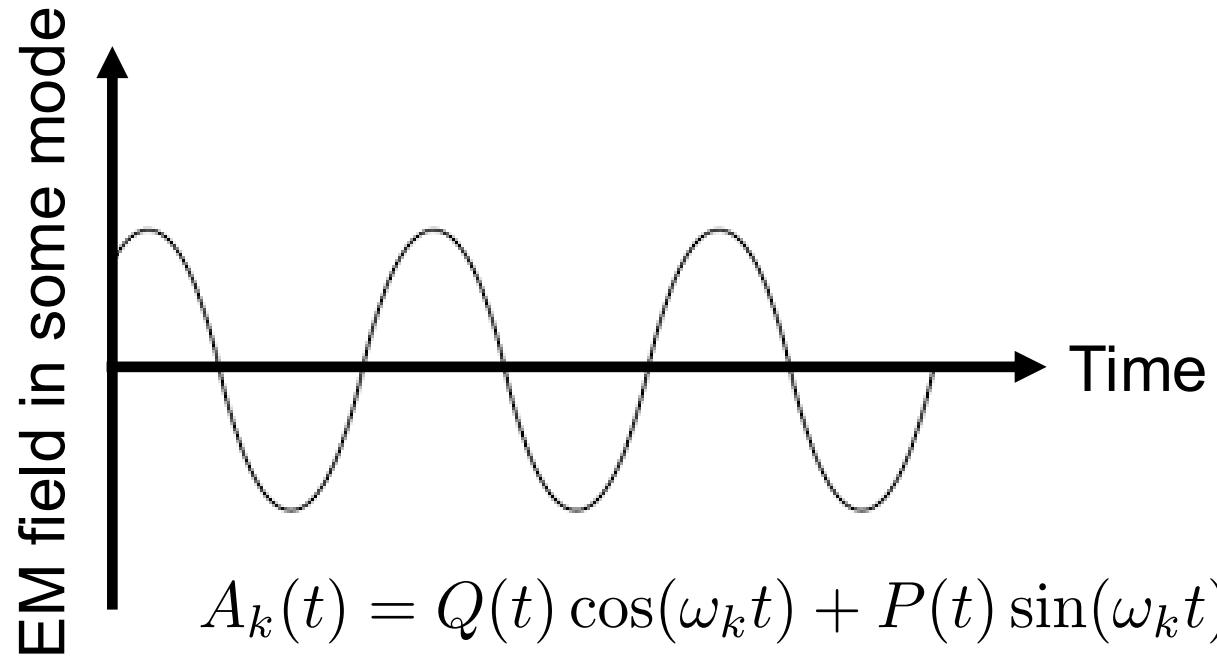


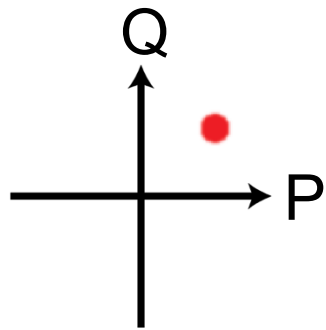
# Controlling quantum fluctuations with nonlinear interactions

Nick Rivera (PhD '22, MIT Physics)  
Junior Fellow, Harvard Society of Fellows  
Department of Physics, Harvard University

# Quantum fluctuations in electromagnetic fields

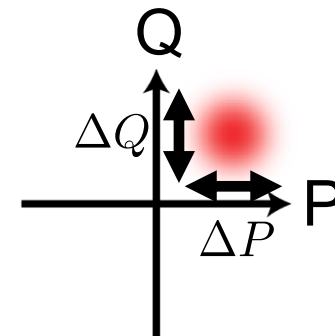


Classical states



Q, P can be definite

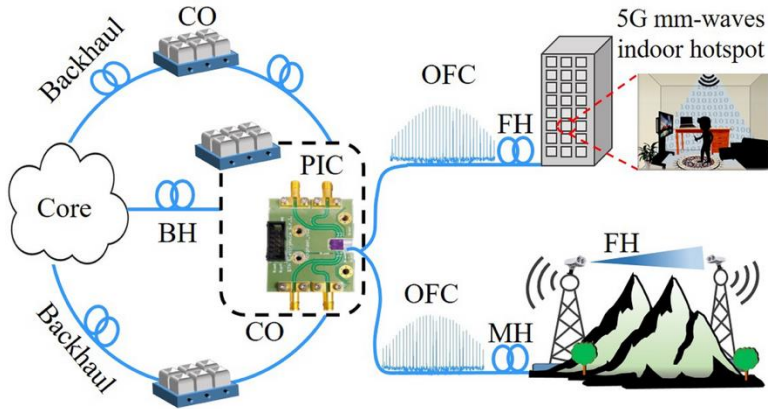
Quantum states: uncertainty



$$\Delta Q \Delta P \geq 1$$

# Limits from quantum noise in different domains

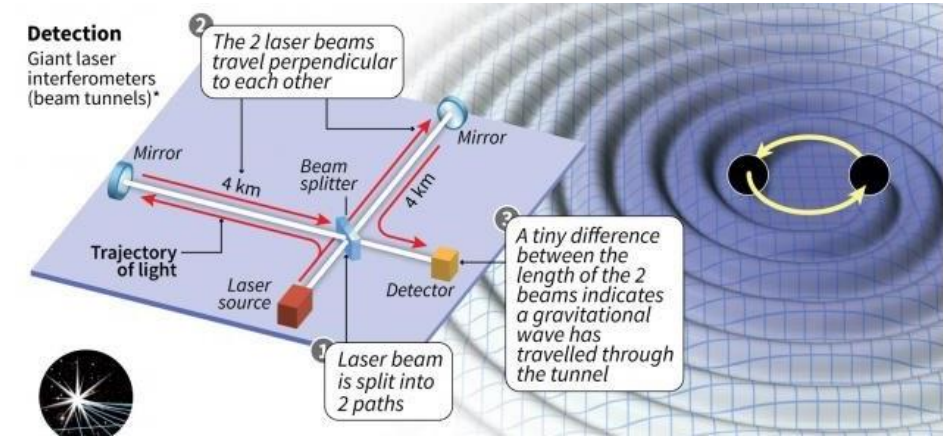
## Systems limited by quantum and similar noises



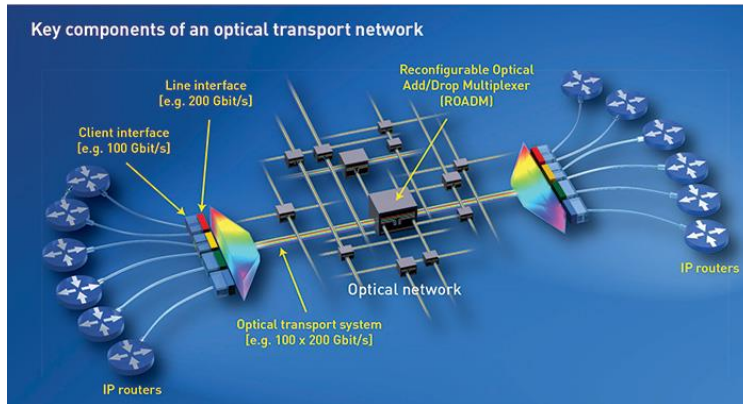
5G/6G networks



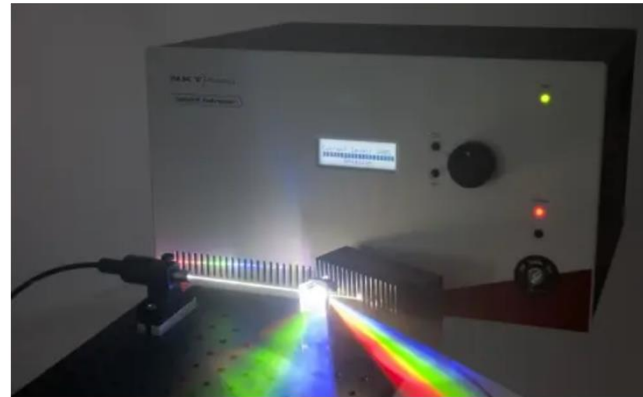
Medicine



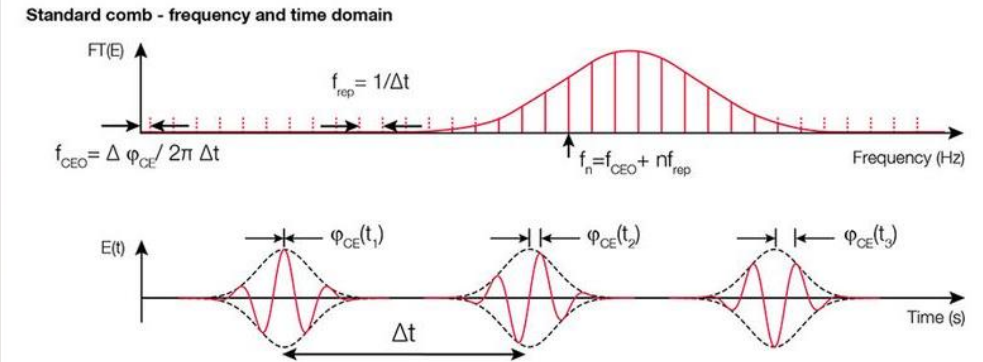
Interferometers



Optical fiber networks

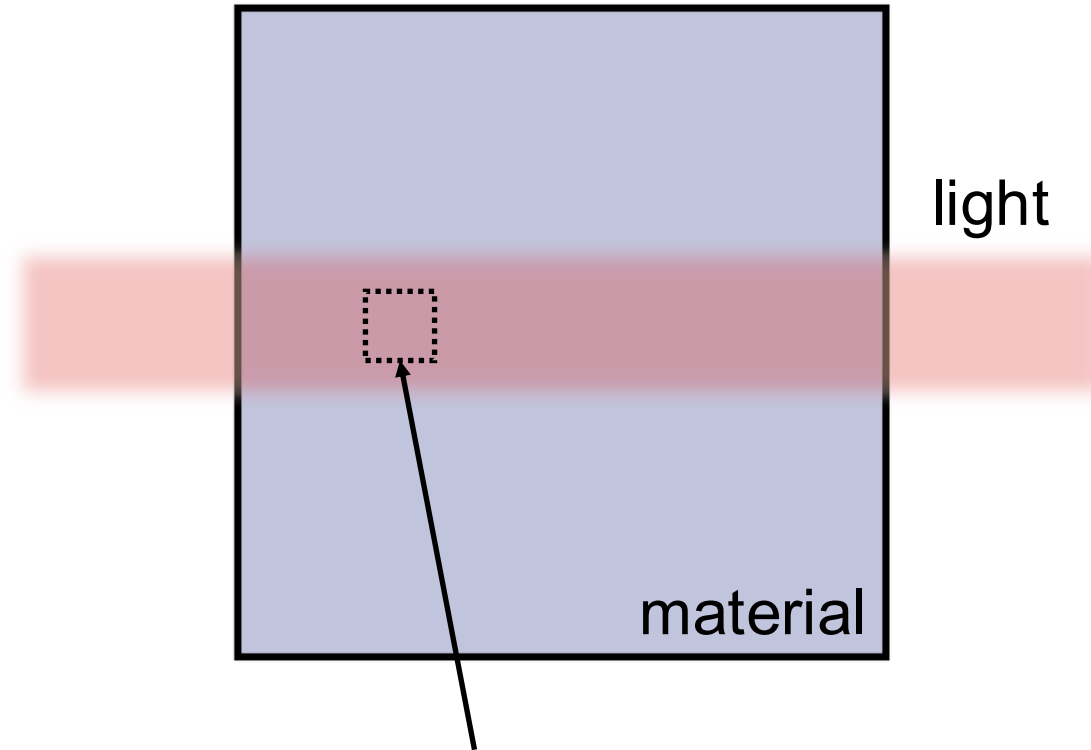


Light sources



Precision measurement

# Nonlinear response of matter to light



$$P_i = \epsilon_0 \left( \underbrace{\chi_{ij}^{(1)} E_j}_{\text{Weak fields}} + \underbrace{\chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \dots}_{\text{Stronger driving fields (e.g., using focused lasers)}} \right)$$

Material  
polarization

Weak  
fields

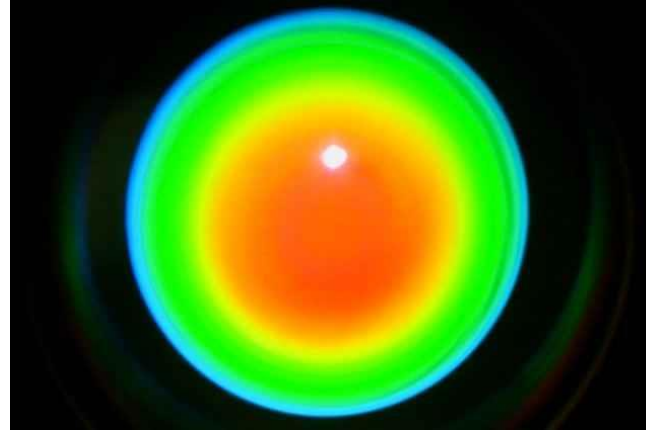
Stronger driving fields  
(e.g., using focused lasers)

# Manifestations of nonlinear optics

Harmonic generation



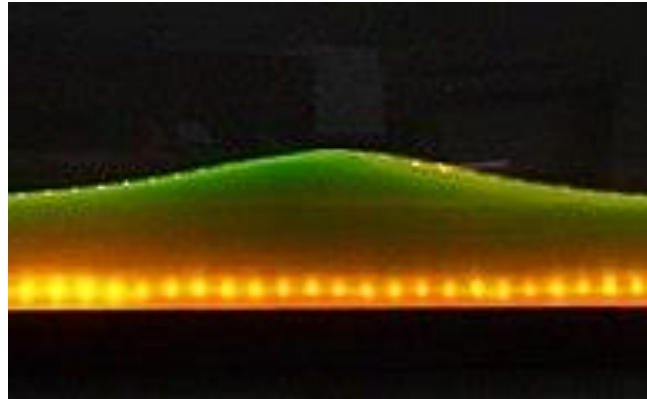
Frequency down-conversion



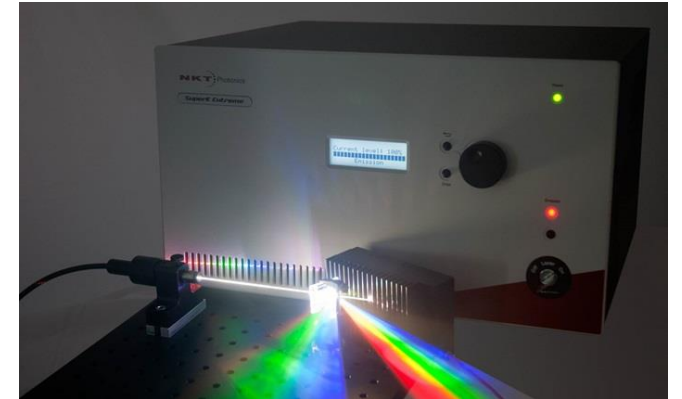
Electro-optic effect



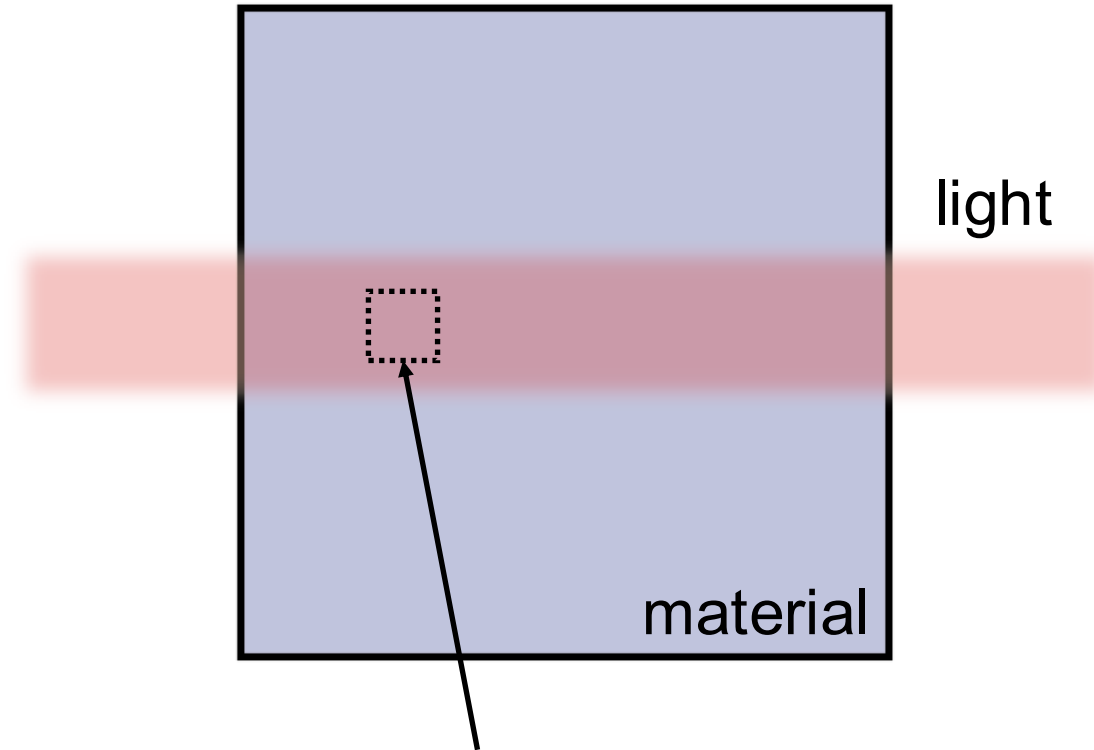
Solitons of light



Supercontinuum generation



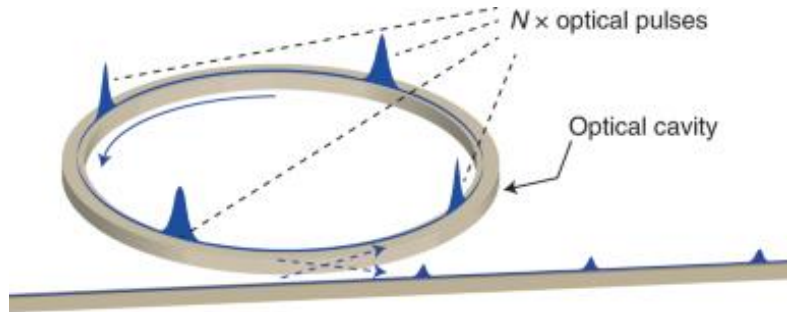
# The quantum side of nonlinear optics



$$H = \int d\mathbf{r} \frac{1}{2} \left( \underbrace{\epsilon E^2 + \mu H^2}_{\text{linear optics}} + \underbrace{\eta^{(2)} E^3 + \eta^{(3)} E^4 + \dots}_{\text{photon-photon interactions: quantum states of light like "squeezed states"}} \right)$$

# Motivation: quantum nature of complex nonlinear systems

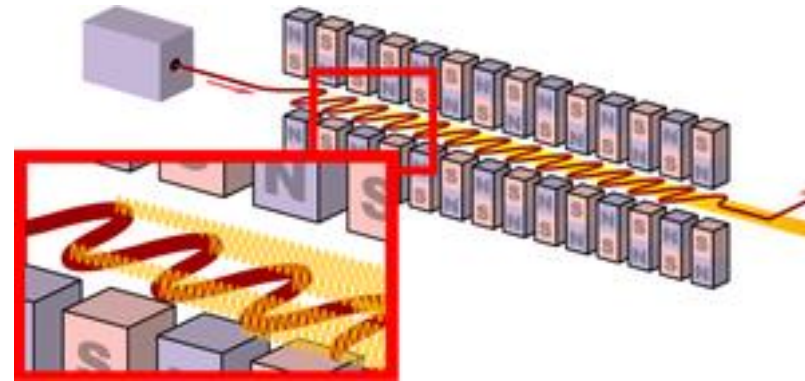
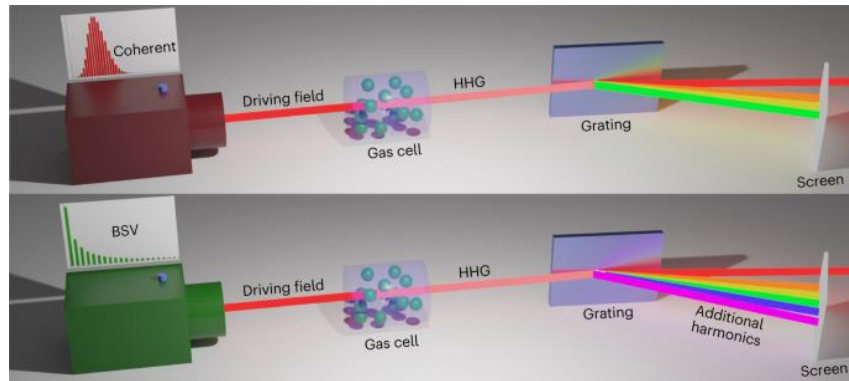
Frequency combs: dissipative solitons



Supercontinuum generation



X-ray sources: High-harmonic generation and free-electron lasers



Huge range of phenomena which we understand classically but know little about the quantum optical properties (correlations, noise, etc.) relevant to many current research problems. No framework that directly leverages mature classical understanding!



Shiekh Zia Uddin  
(MIT)

# The quantum optics of supercontinuum generation

Based on: Uddin\* and **Rivera\*** *et al.* arXiv:2311.05535 (2023)

# Supercontinuum generation



“White as a lamp, bright as a laser”

# The classical description of supercontinuum generation

incident fs pulse:  $A(z,t)$

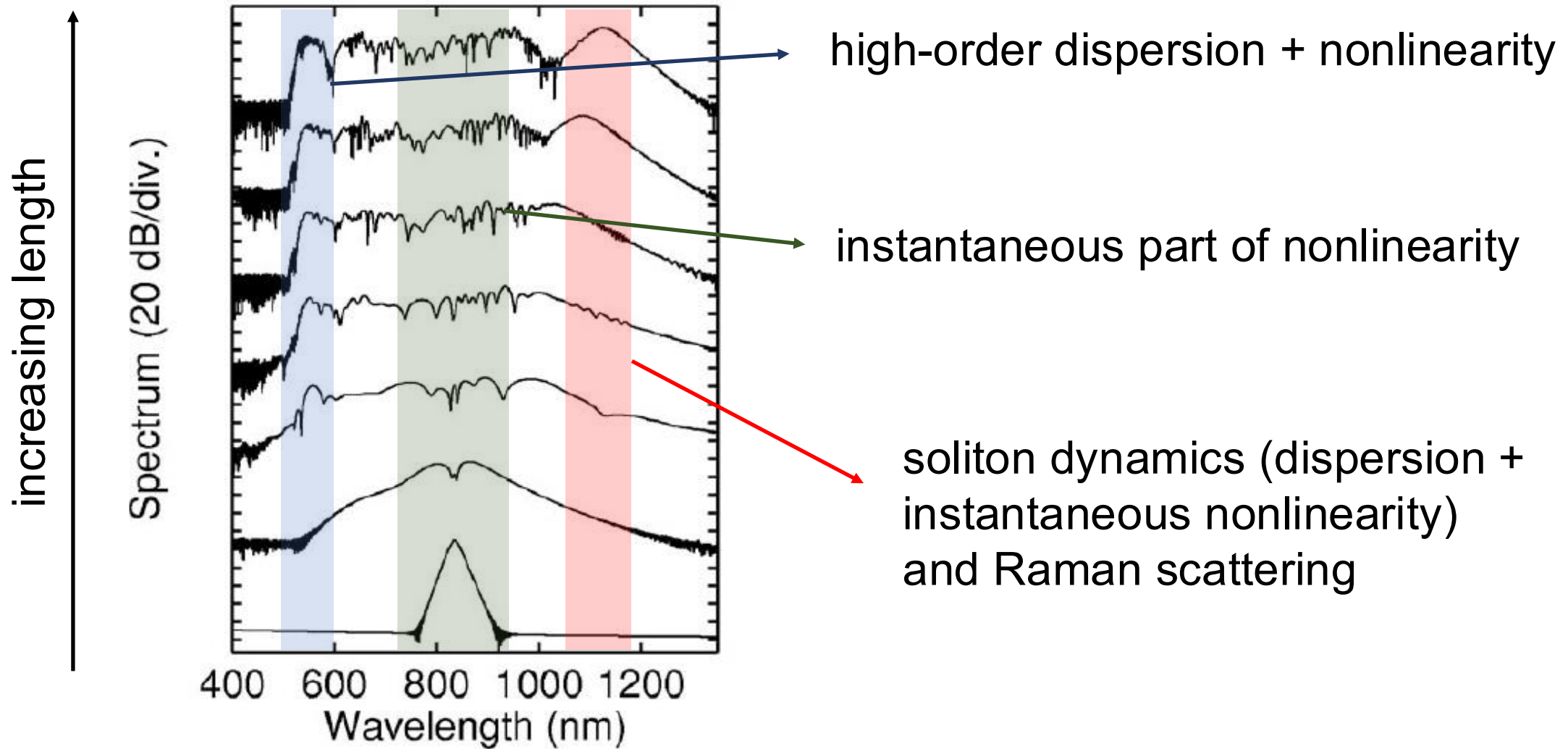


**Pulse evolution: the generalized nonlinear Schrodinger equation**

$$\partial_z A(z, t) = \underbrace{-\frac{\alpha}{2} A(z, t)}_{\text{loss}} + \underbrace{\sum_{k=2}^{\infty} \frac{i^{k+1} \beta_k}{k!} \partial_t^k A(z, t)}_{\text{dispersion: } k = k(\omega)} + \underbrace{i\gamma A(z, t) \int dt' R(t') |A(z, t - t')|^2}_{\text{Nonlinearity: instantaneous (electronic) + delayed (phonons)}}$$

This equation (+ variants) has been studied by many authors for a long time (50+ years)!

# The physical processes leading to supercontinuum generation



Dudley *et al.* *Rev. Mod. Phys.* (2006).

# Quantum fluctuations in supercontinuum generation

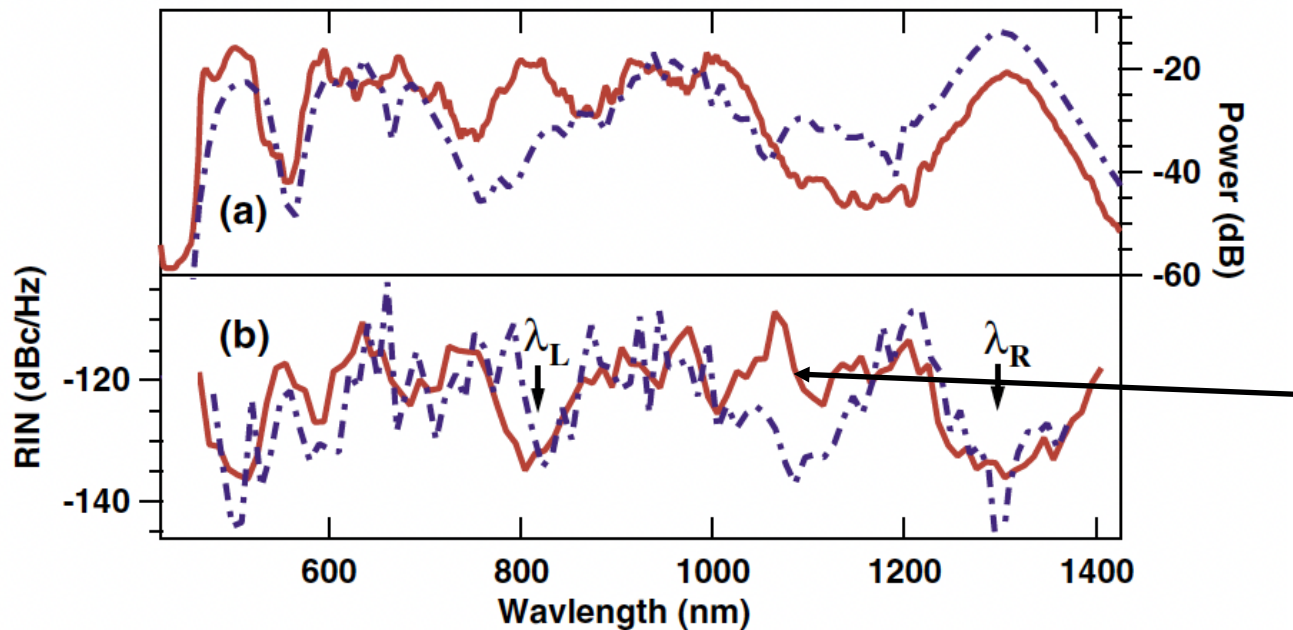
VOLUME 90, NUMBER 11

PHYSICAL REVIEW LETTERS

week ending  
21 MARCH 2003

## Fundamental Noise Limitations to Supercontinuum Generation in Microstructure Fiber

K. L. Corwin,<sup>1</sup> N. R. Newbury,<sup>1</sup> J. M. Dudley,<sup>2</sup> S. Coen,<sup>3</sup> S. A. Diddams,<sup>1</sup> K. Weber,<sup>1</sup> and R. S. Windeler<sup>4</sup>

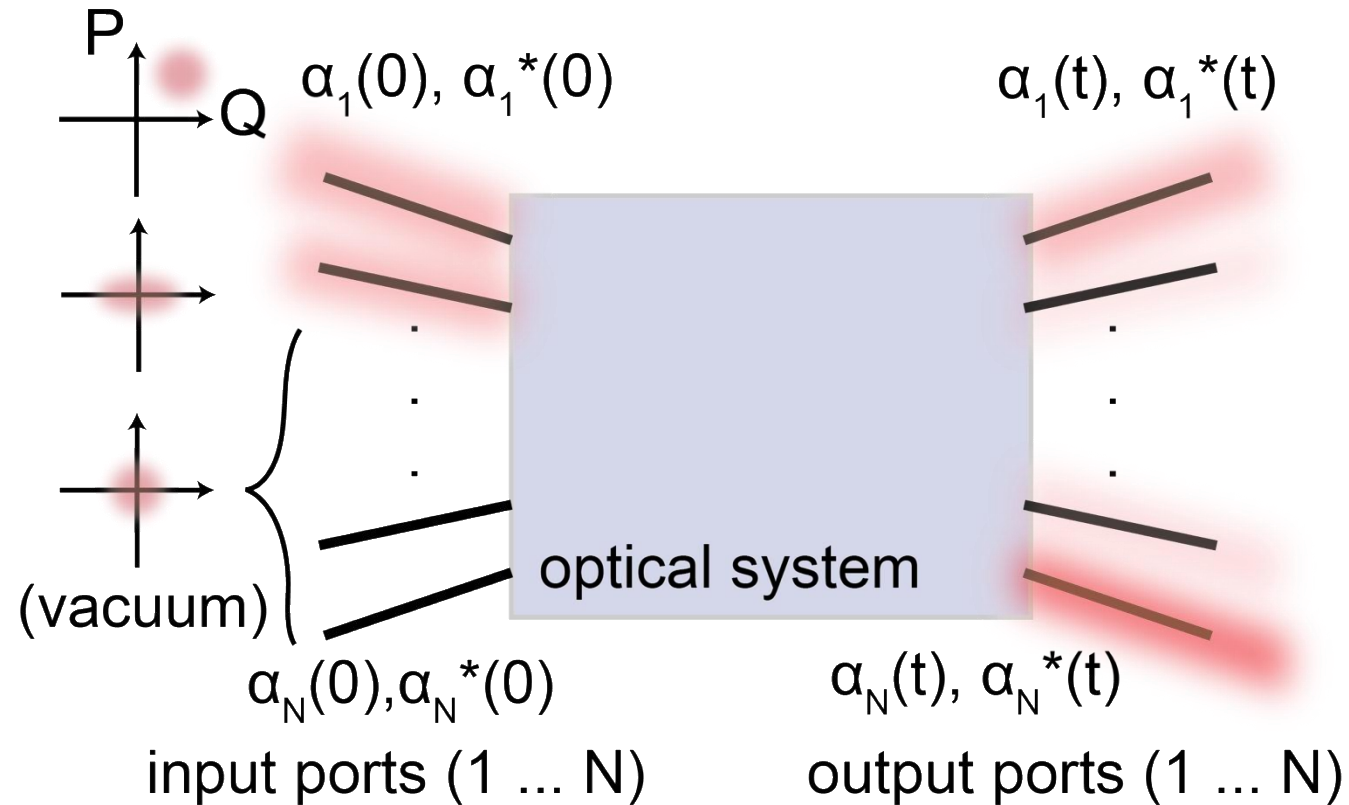


Nearly 1-10% pulse-to-pulse fluctuation from inputs with <0.01%. Individual wavelengths are not in coherent states, even when initial light is!

Corwin *et al.* PRL (2003).

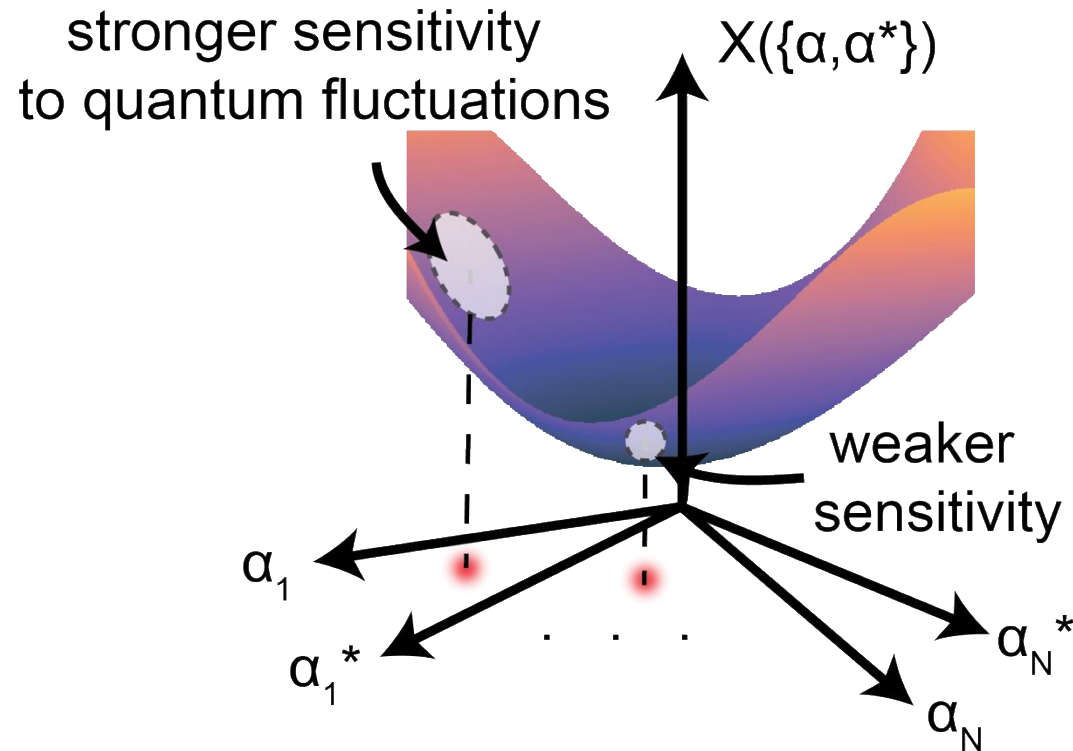
**Picture of supercontinuum: fundamentally noisy and noise-sensitive.**

# Quantum noise in nonlinear dynamical systems



Input-output picture of classical dynamics:  $\alpha_i(t) = F[\{\alpha_j(0), \alpha_j^*(0)\}]$

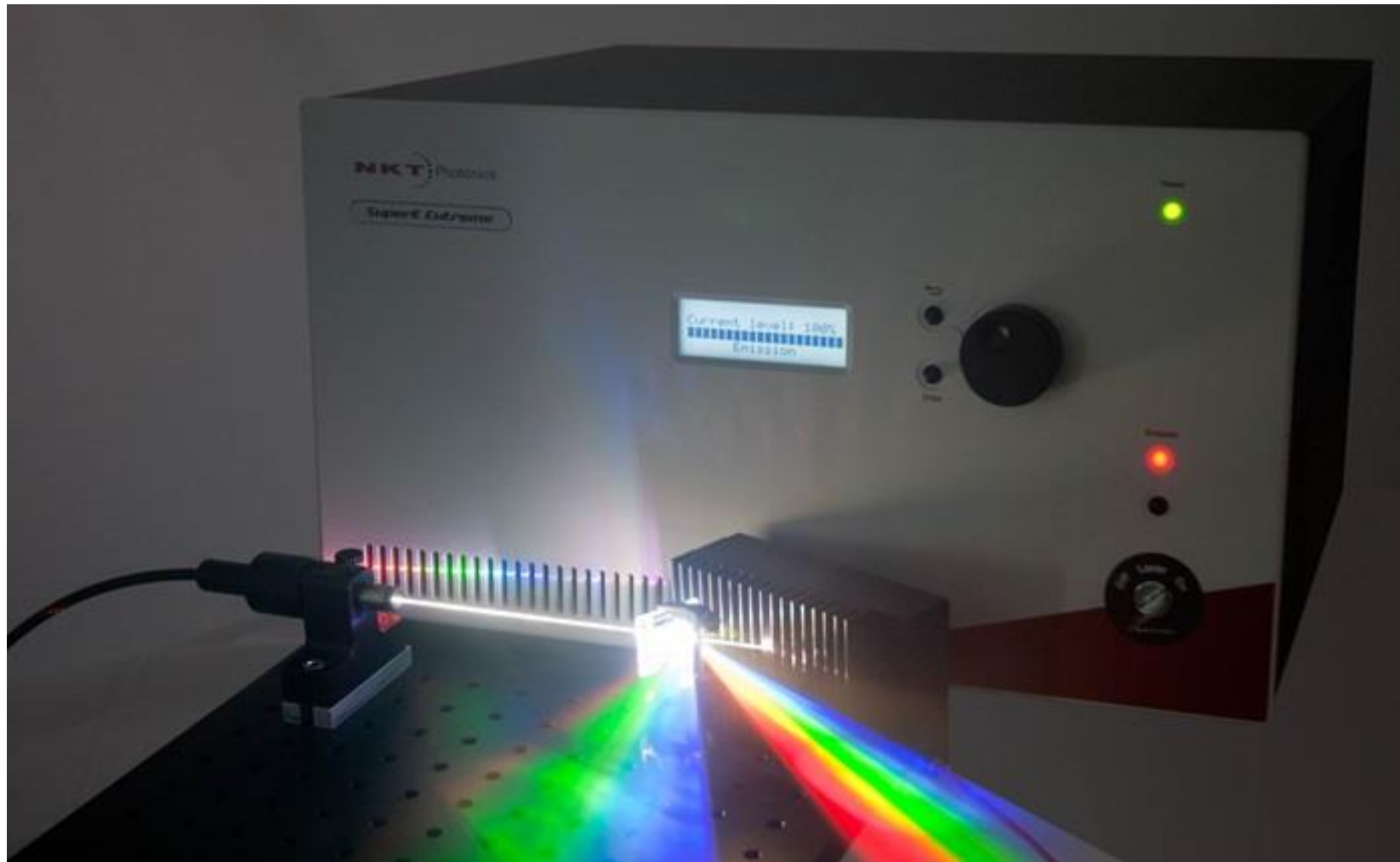
# Quantum sensitivity analysis of nonlinear dynamics



**Crux:** analyze response of observable to quantum or other noise in the initial conditions of the classical theory. Generally, noise of observable  $X$  given by:

$$(\Delta X(t))^2 = \begin{pmatrix} \frac{\partial X(t)}{\partial \alpha(0)} & \frac{\partial X(t)}{\partial \alpha^*(0)} \end{pmatrix}^T \begin{pmatrix} \langle \delta \alpha \delta \alpha \rangle & \langle \delta \alpha \delta \alpha^* \rangle \\ \langle \delta \alpha^* \delta \alpha \rangle & \langle \delta \alpha^* \delta \alpha^* \rangle \end{pmatrix} \begin{pmatrix} \frac{\partial X(t)}{\partial \alpha(0)} & \frac{\partial X(t)}{\partial \alpha^*(0)} \end{pmatrix}$$

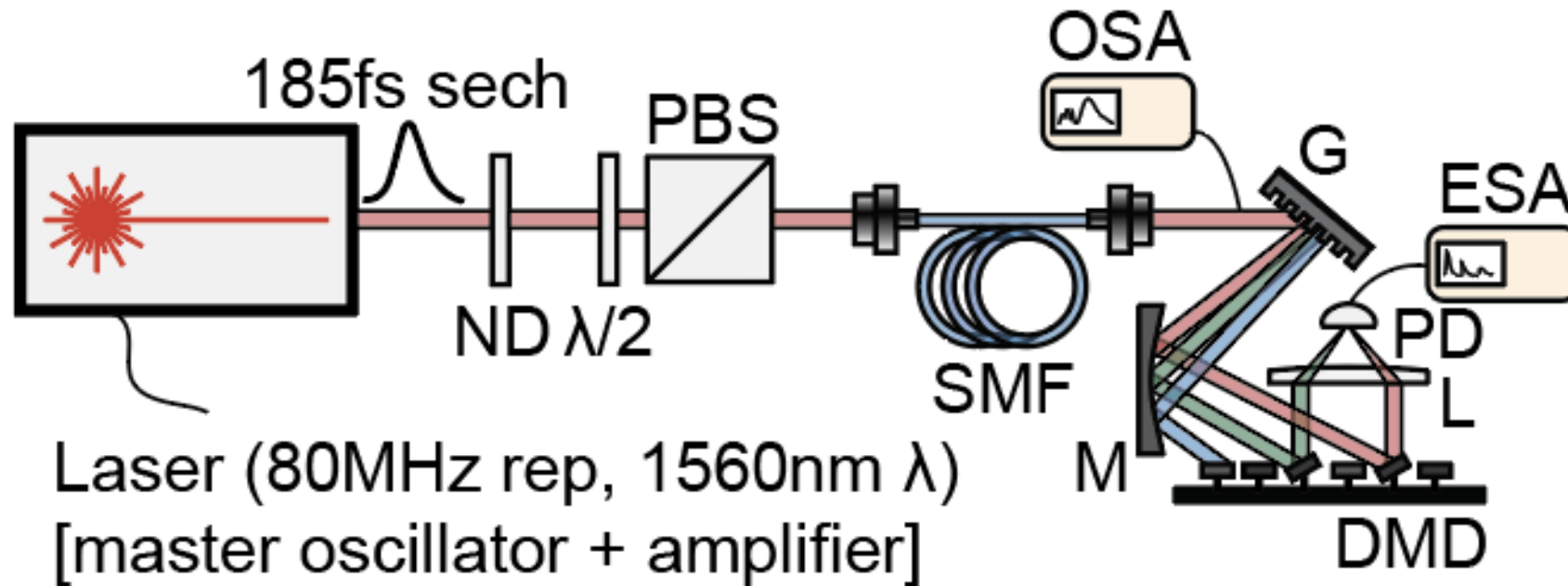
# Quantum optics of supercontinuum generation



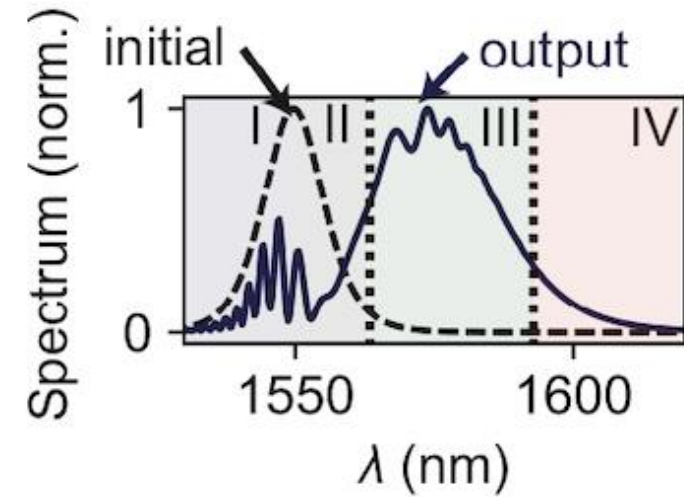
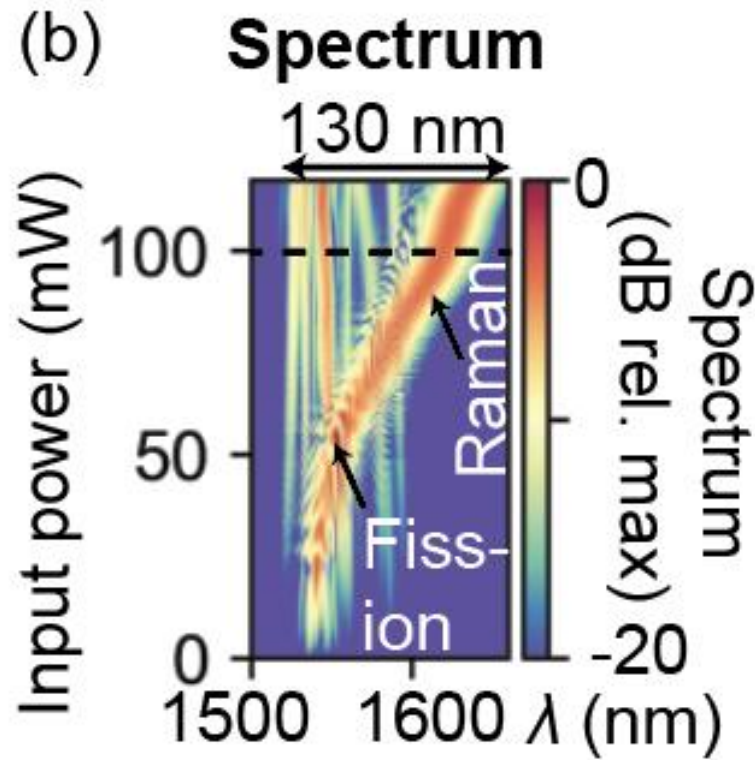
“White as a lamp, bright as a laser”

# A new low noise regime in supercontinuum generation

(a) Experimental schematic

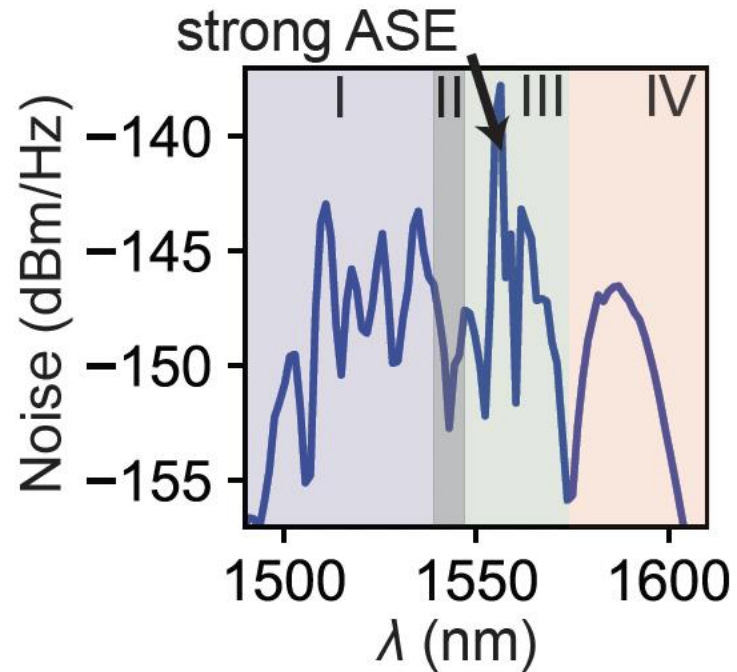


# A new low noise regime in supercontinuum generation

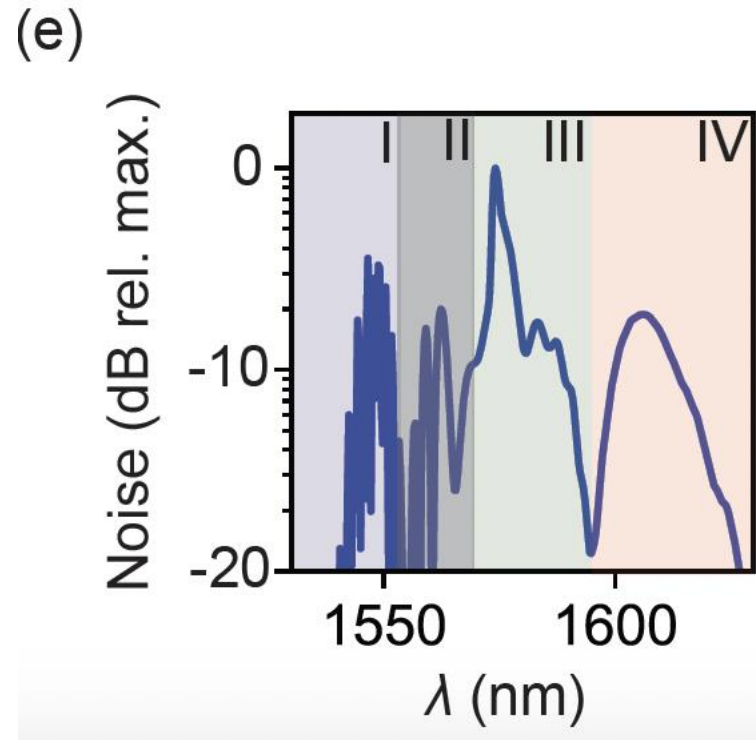
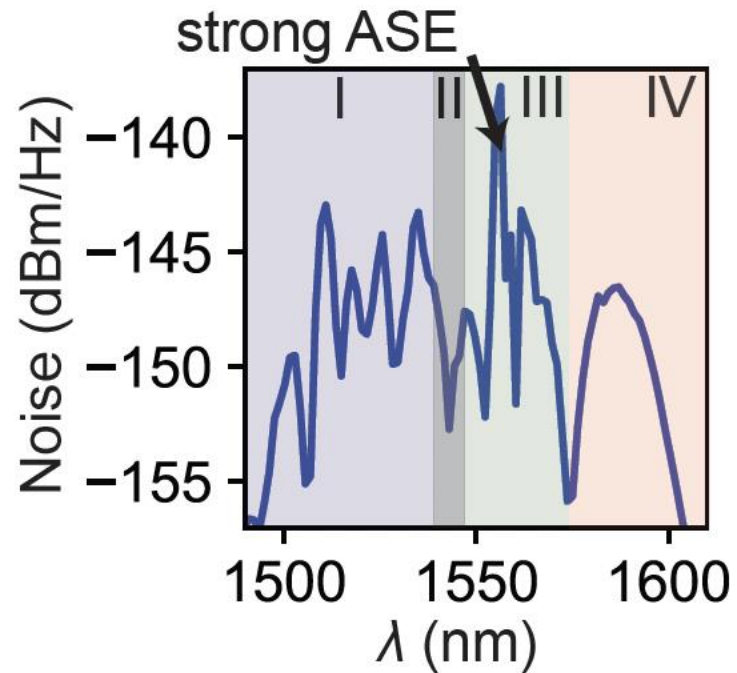


- I: SPM + dispersive waves
- II: SPM + Raman sol. (blue)
- III: Raman sol. (peak)
- IV: Raman sol. (red)

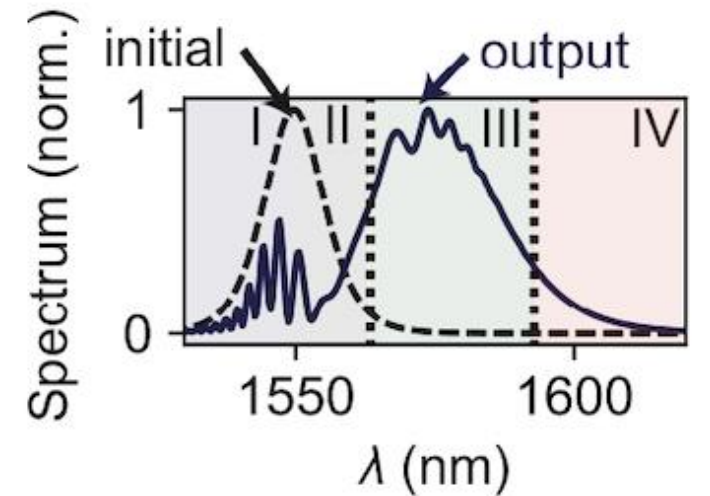
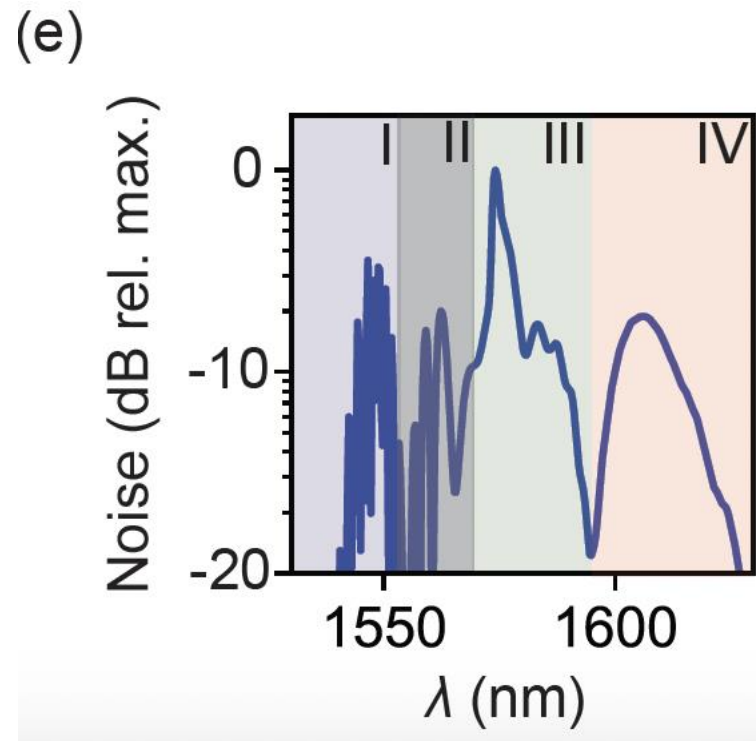
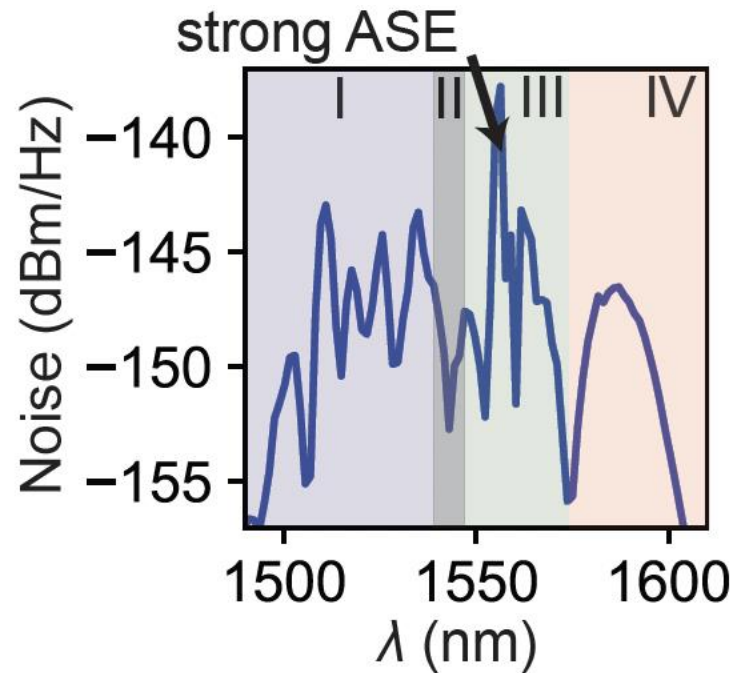
# A new low noise regime in supercontinuum generation



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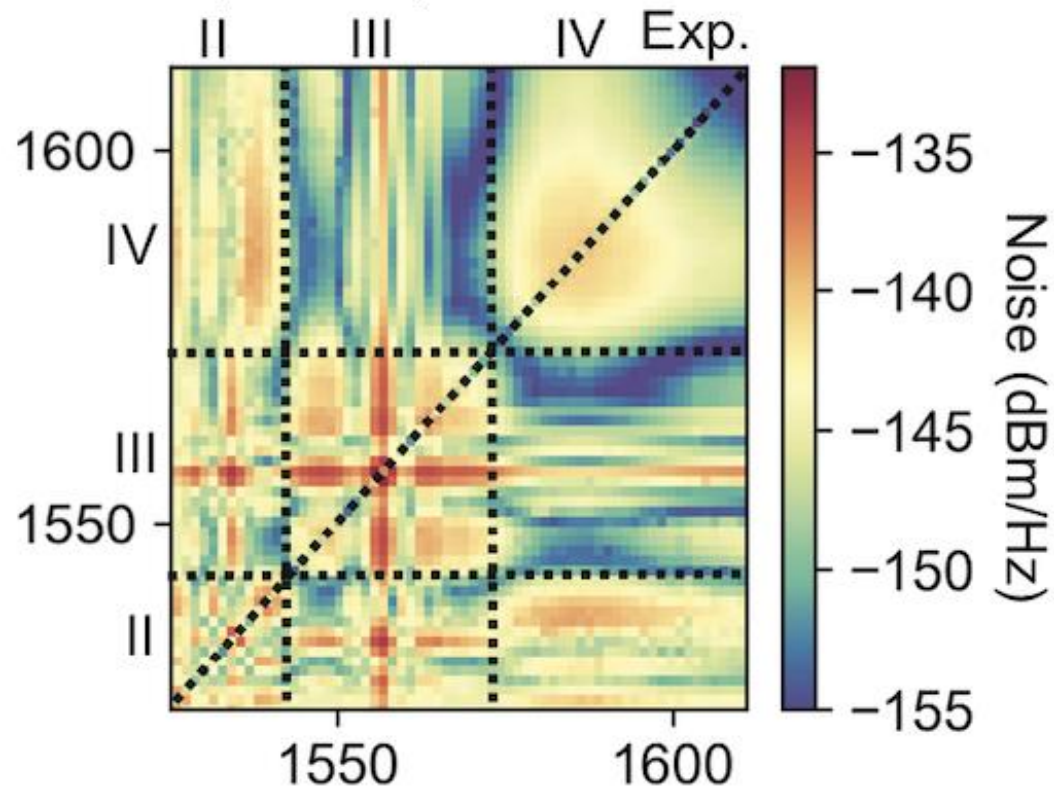


# A new low noise regime in supercontinuum generation

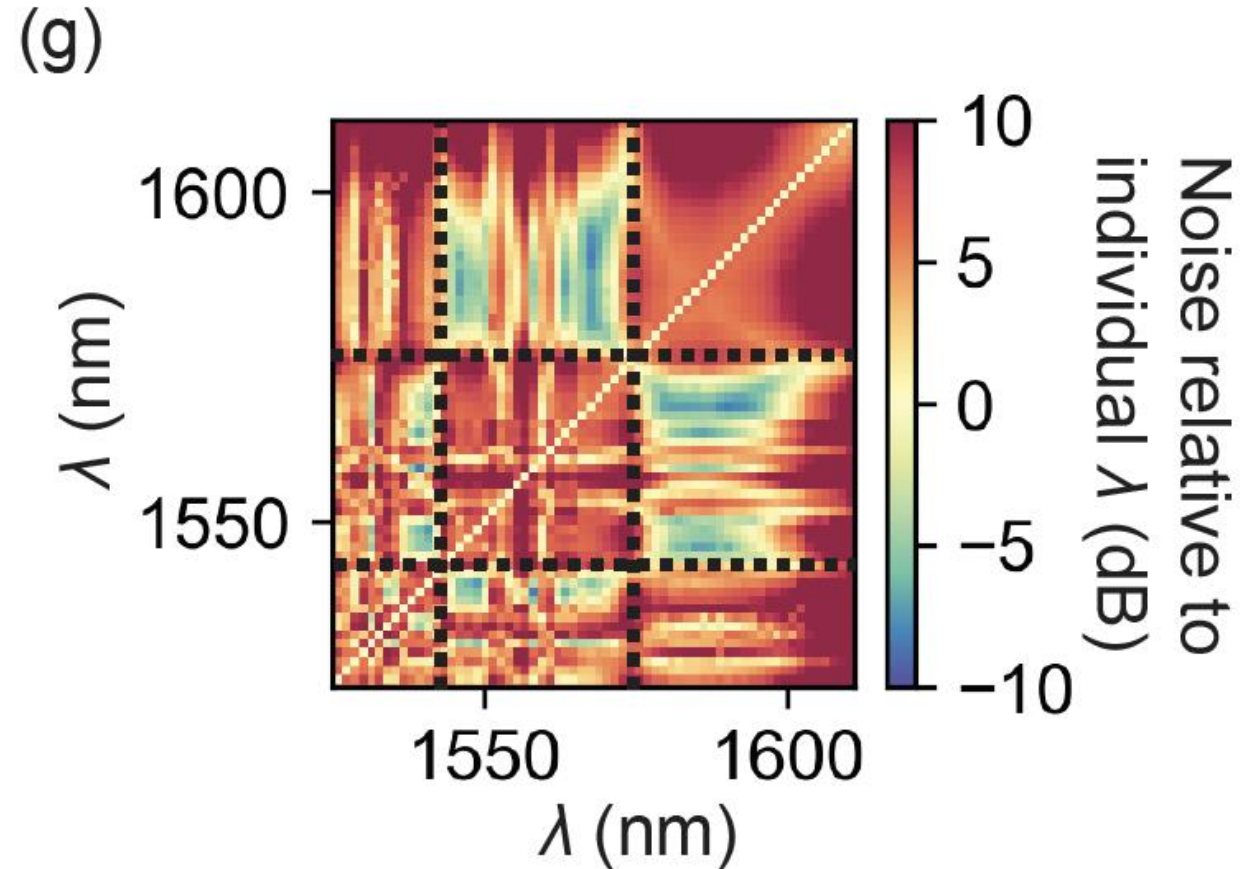
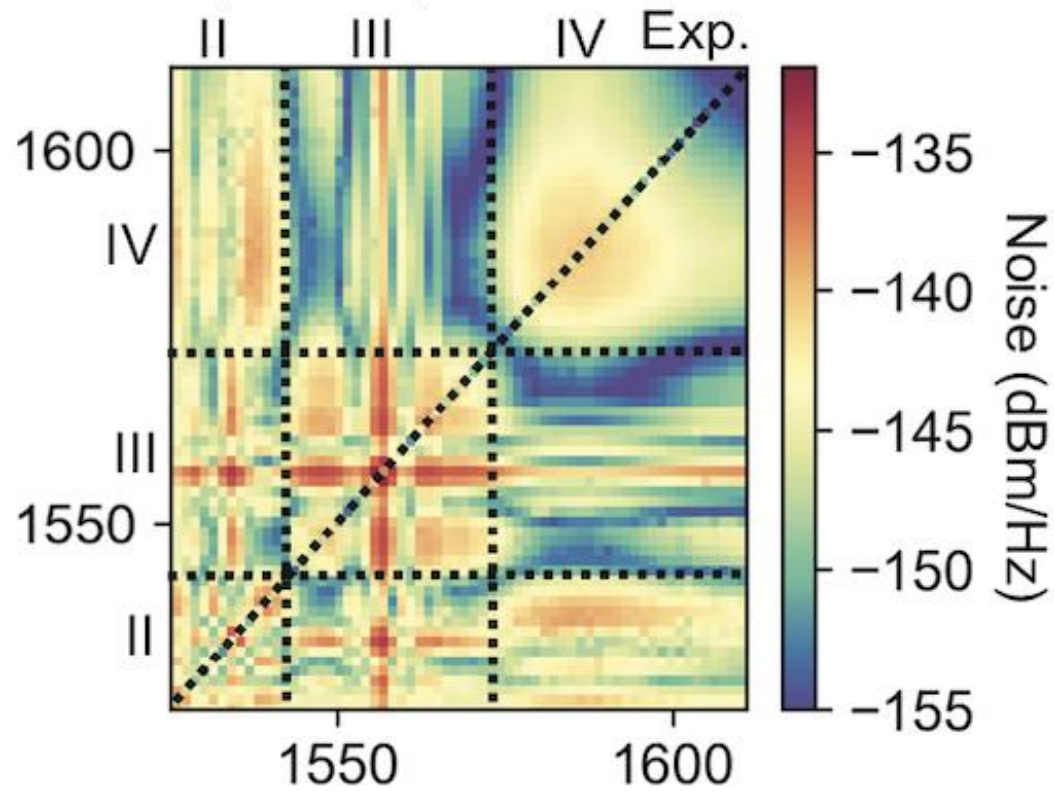


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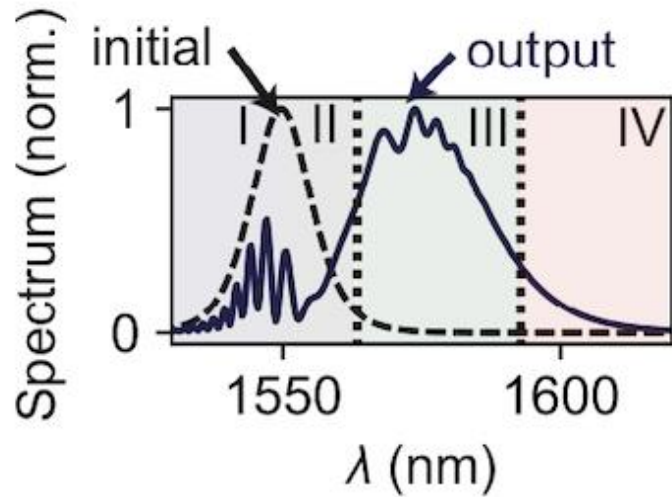
# Low noise multi-wavelength states in supercontinuum



# Low noise multi-wavelength states in supercontinuum

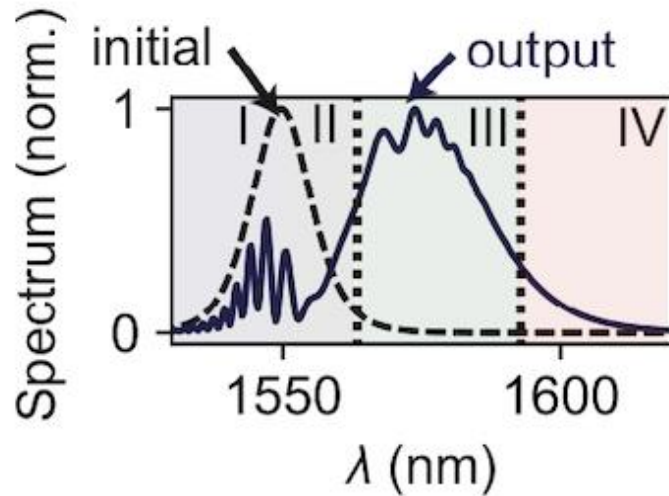


# Low noise multi-wavelength states in supercontinuum



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# Low noise multi-wavelength states in supercontinuum

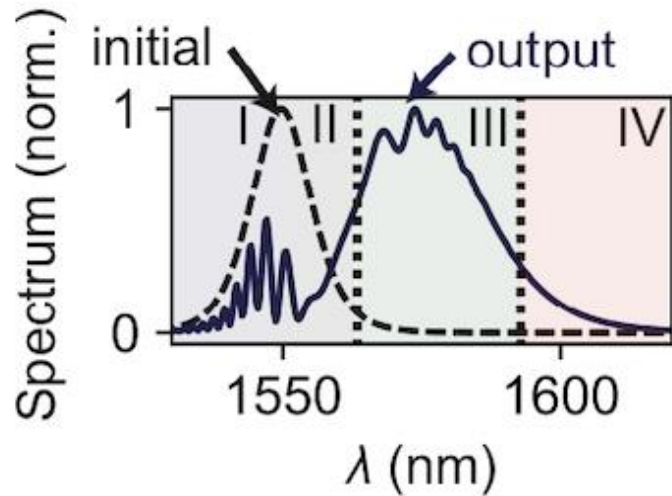


- I: SPM + dispersive waves
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For the example of the low-noise band in supercontinuum, we analyze:

$$\Delta(n_1 + n_2)^2 \equiv \|\nabla_{\alpha(0)}[n_1(t) + n_2(t)]\|^2$$

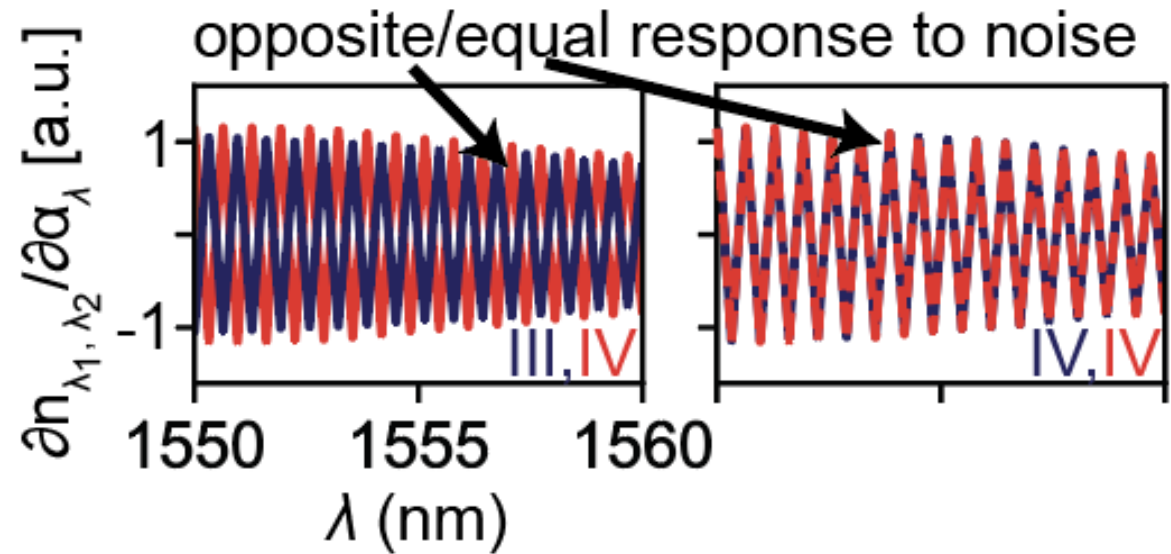
# Low noise multi-wavelength states in supercontinuum



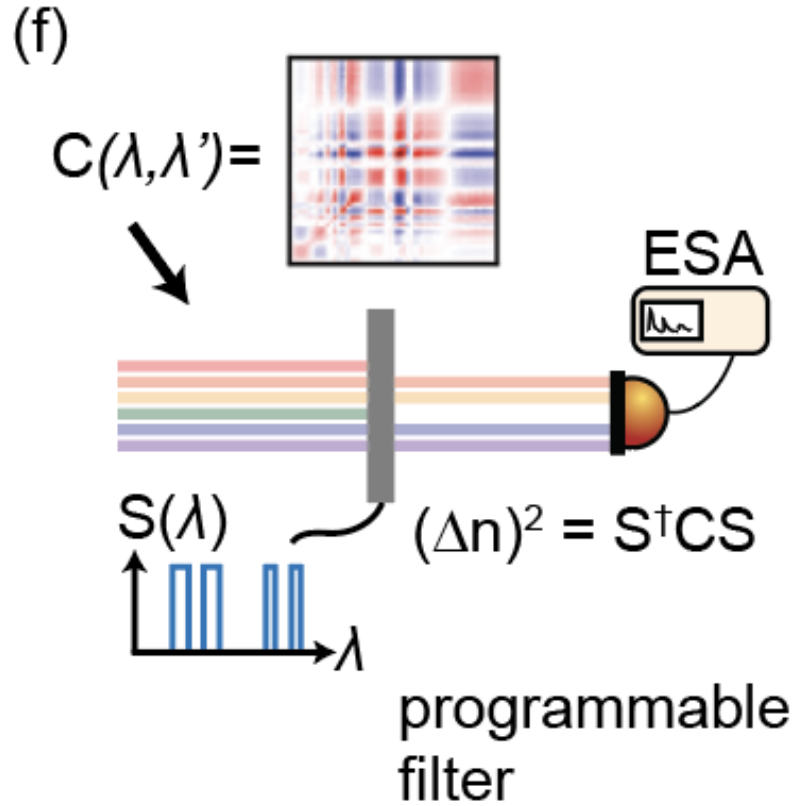
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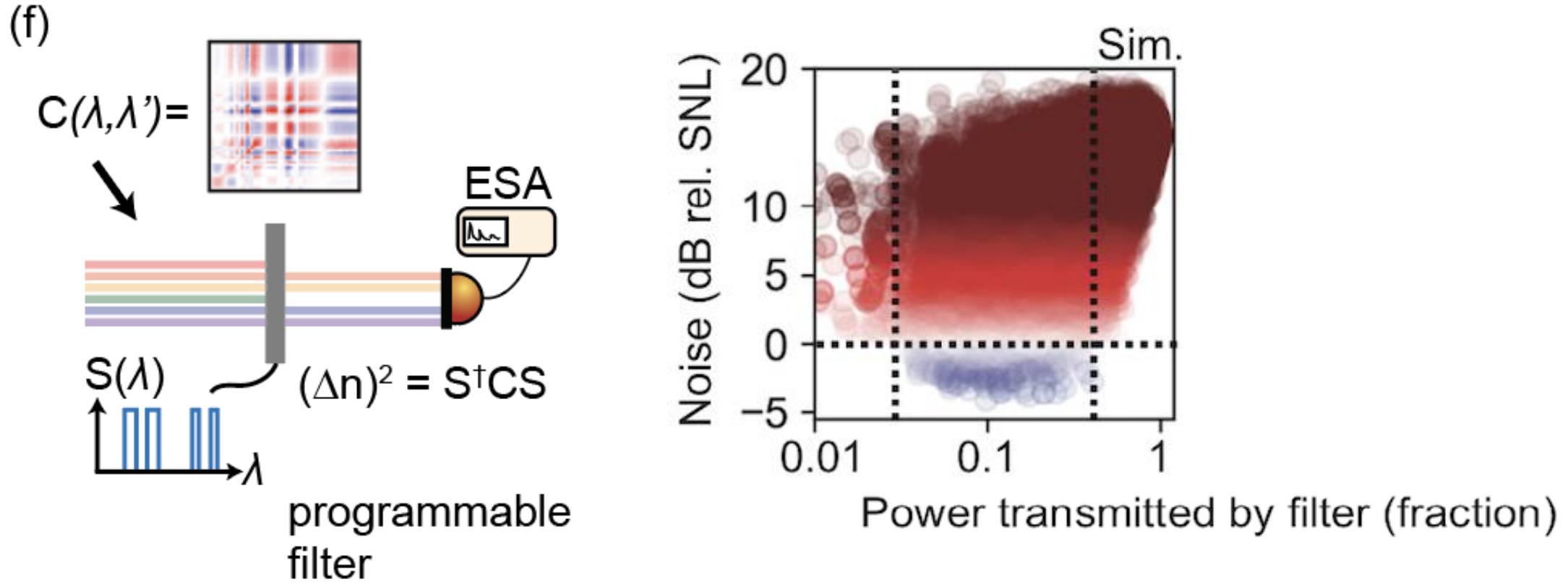
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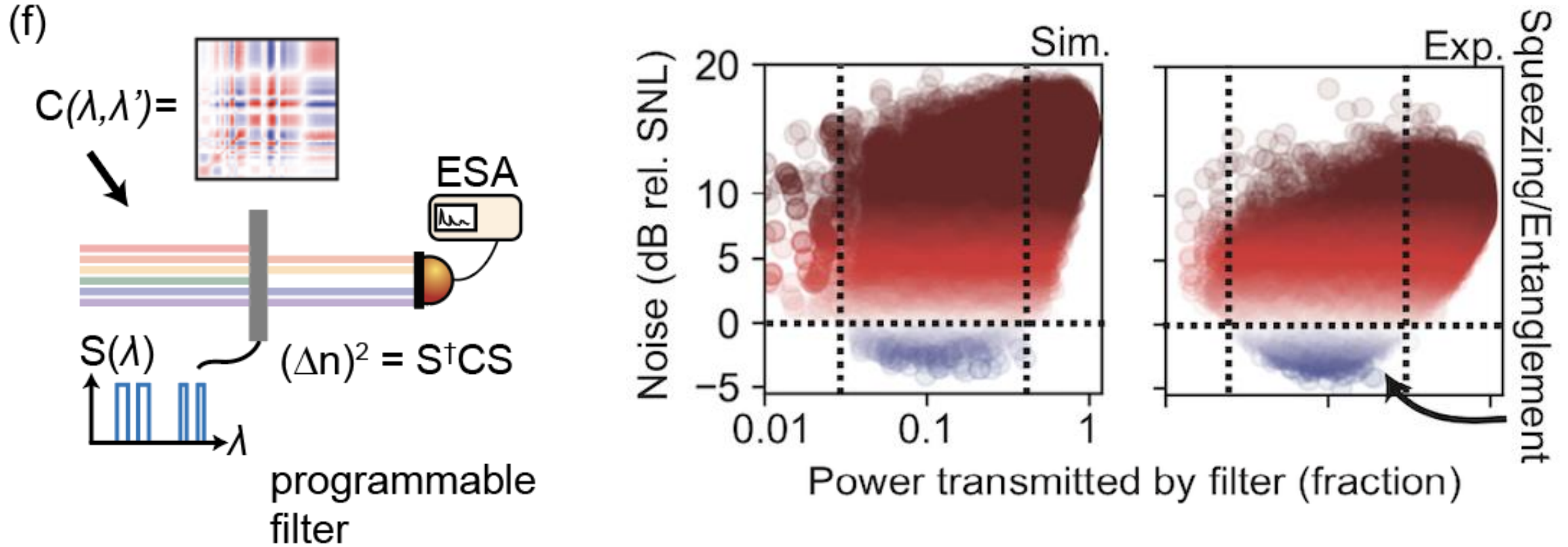
# Squeezed states in supercontinuum



# Squeezed states in supercontinuum

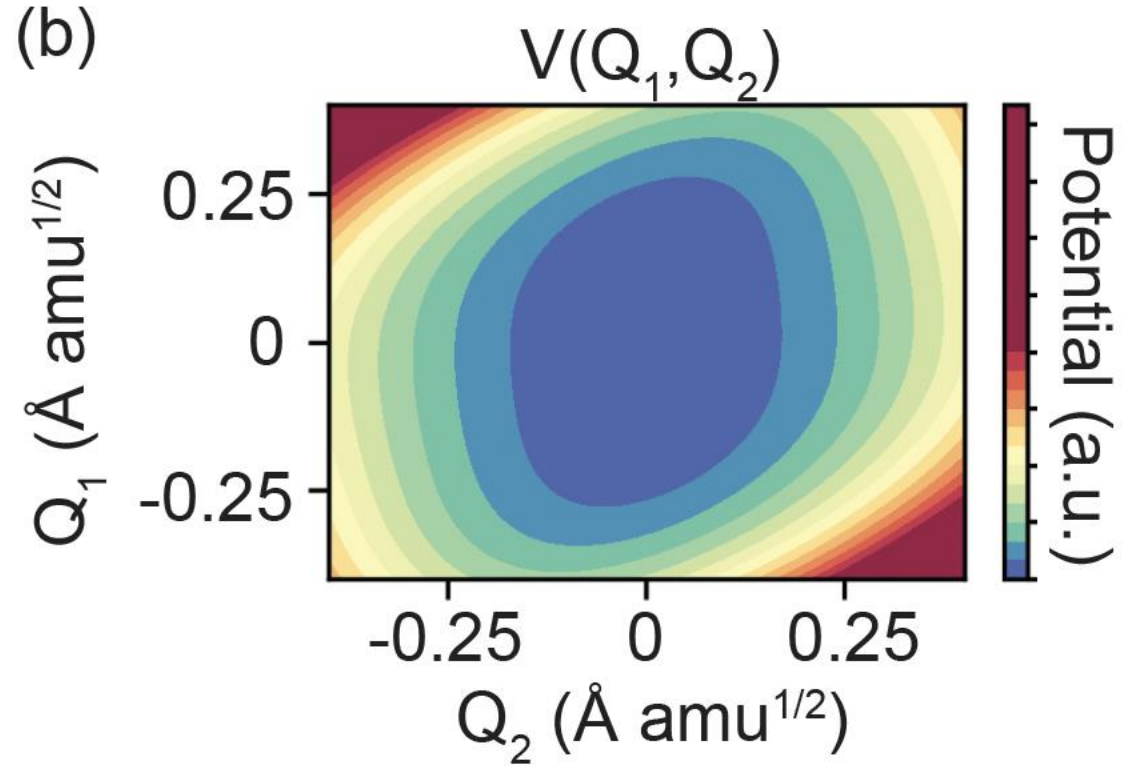
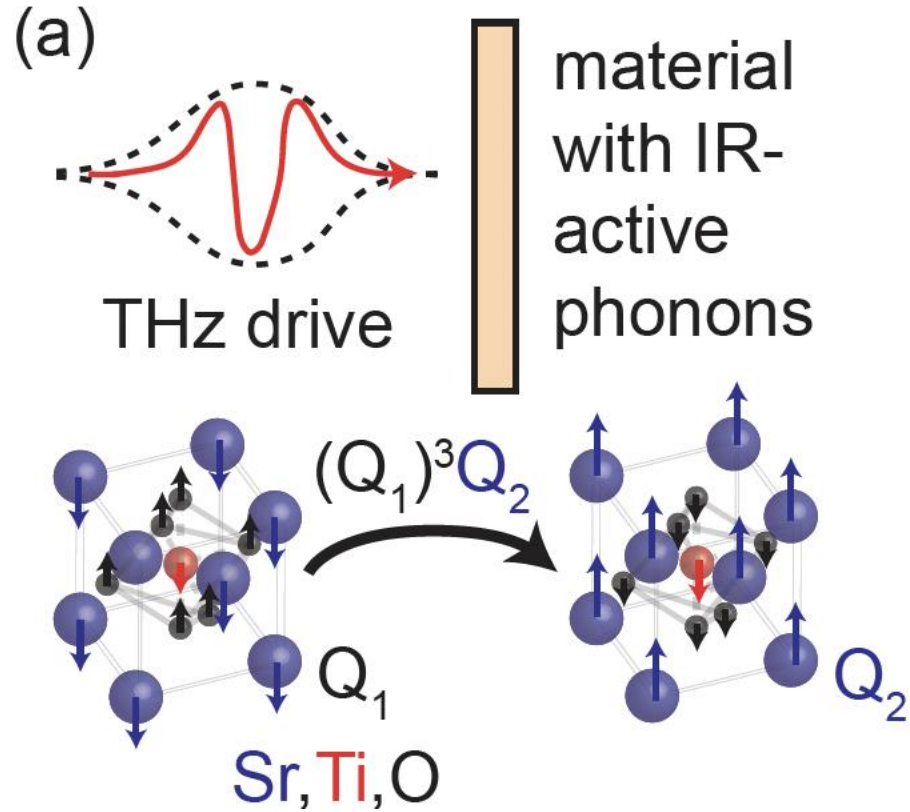


# Squeezed states in supercontinuum



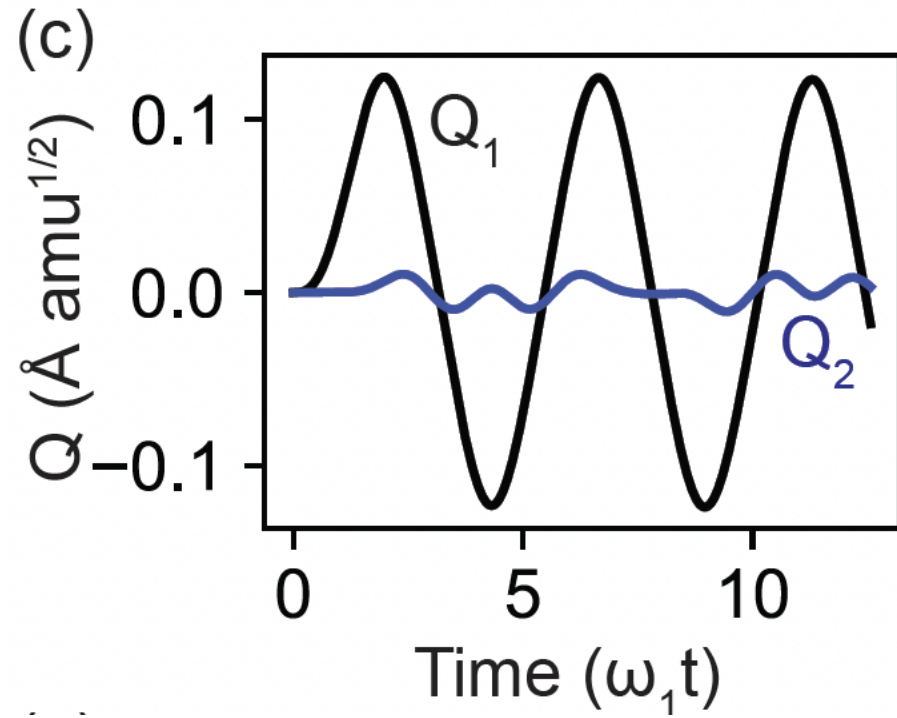
**New directions with this theory**

# Phonon squeezing

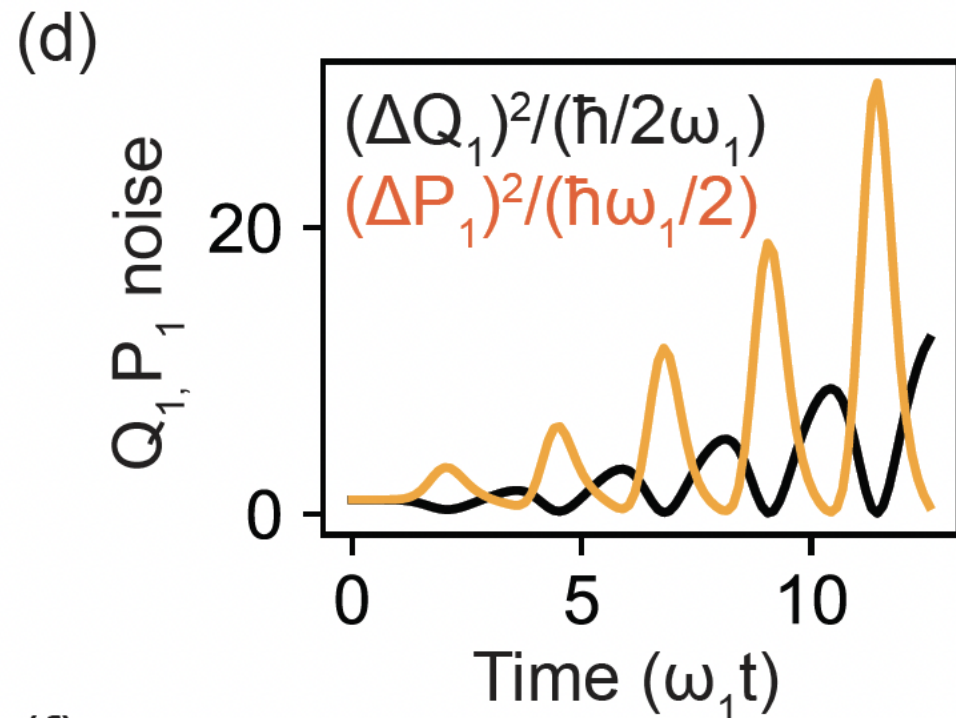
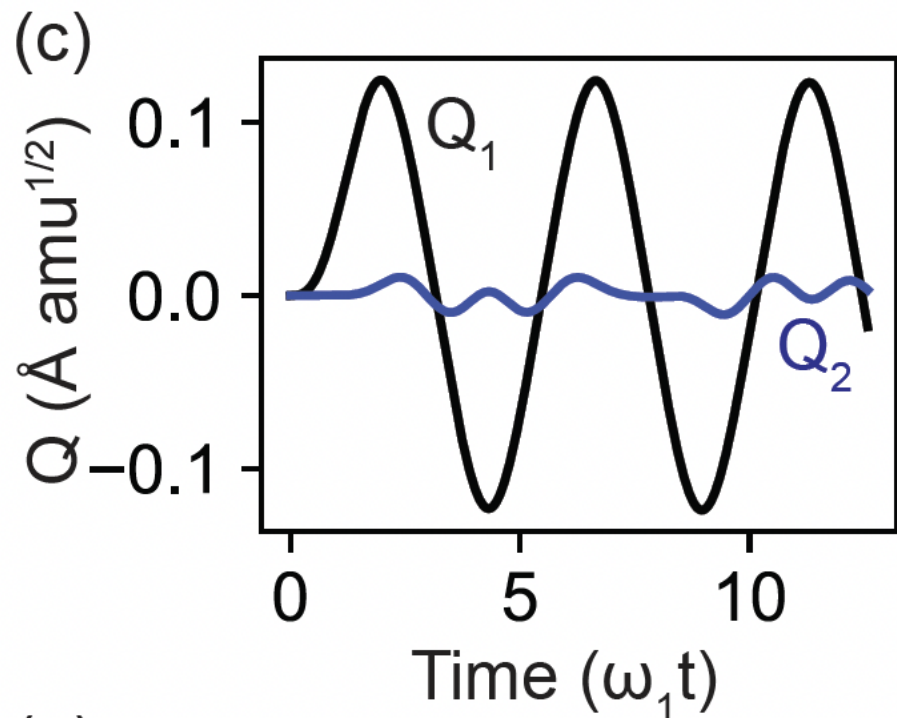


$$V = \frac{1}{2}\omega_1^2 Q_1^2 + \frac{1}{2}\omega_2^2 Q_2^2 + \frac{1}{4}\kappa_1 Q_1^4 + \frac{1}{4}\kappa_2 Q_2^4 + \chi Q_1^2 Q_2^2 + \psi_{12} Q_1^3 Q_2 + \psi_{21} Q_1 Q_2^3$$

# Phonon squeezing

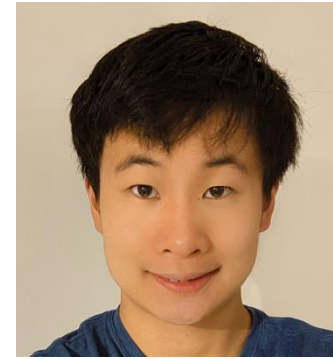


# Phonon squeezing

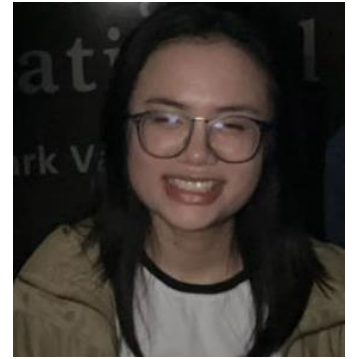




Jamison Sloan



Sean Chen  
(UROP)

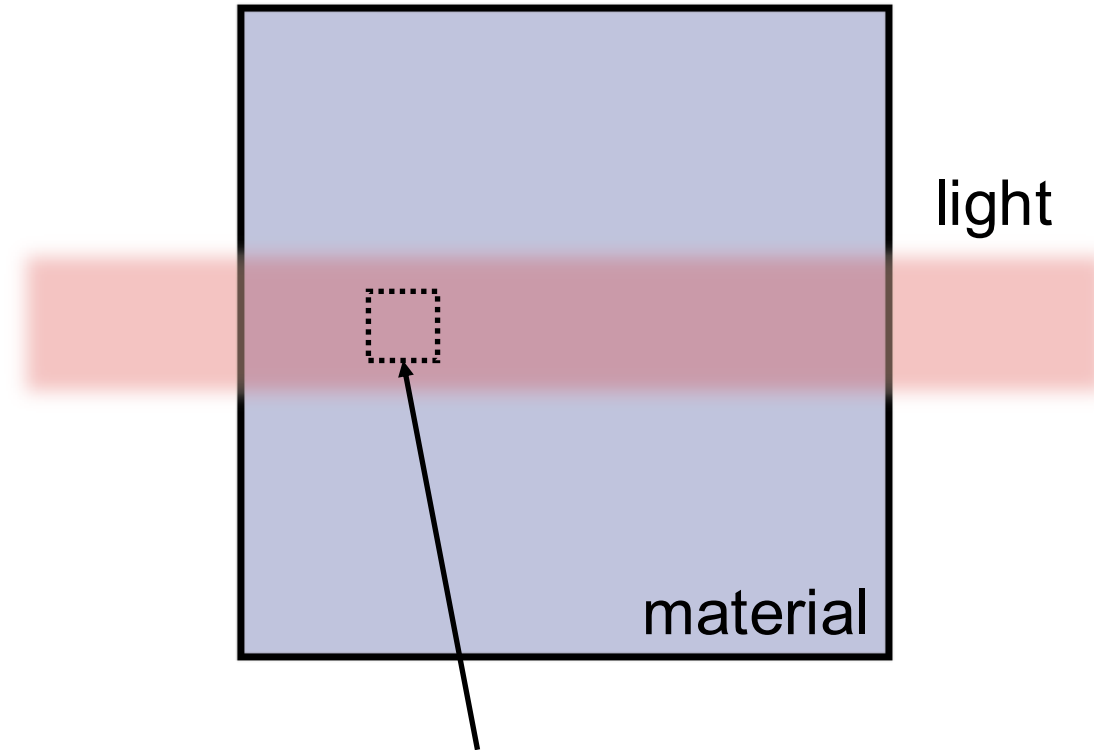


Linh Nguyen  
(UROP)

# Generating fundamental quantum states of light in optics using nonlinear nanophotonics

Based on: **Rivera et al.** PNAS (2023), Chen and **Rivera et al.** arXiv:2312.07386 (2023)

# Changing optical properties with one or few photons



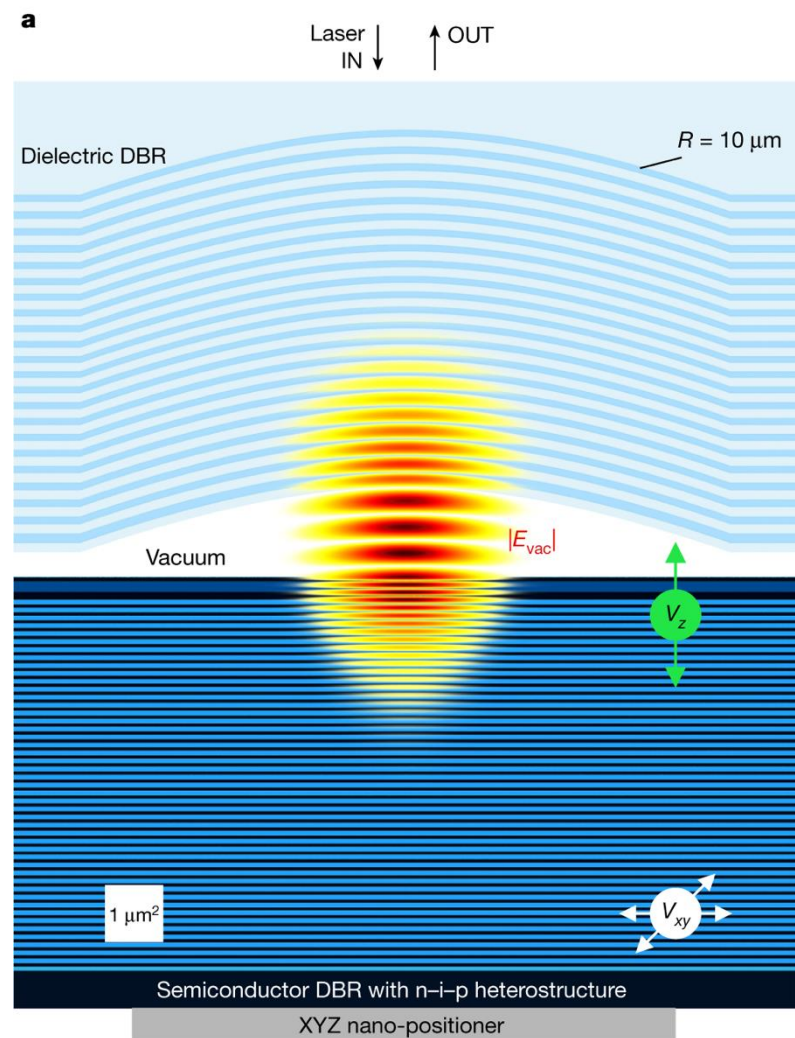
$$P_i = \epsilon_0 \left( \underbrace{\chi_{ij}^{(1)} E_j}_{\text{Material}} + \underbrace{\chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \dots}_{\text{Stronger driving fields}} \right)$$

Material polarization    Weak fields    ~~Stronger driving fields~~  
(e.g., using focused lasers)

**Relevant for "field of a single photon"?**  $E_{\text{SP}} \sim \sqrt{\hbar\omega/2\epsilon_0 V}$

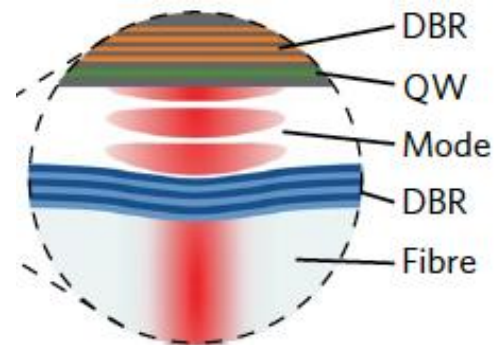
# Systems for realizing nonlinear *optics* at few-photon scales

## Two-level systems in cavities



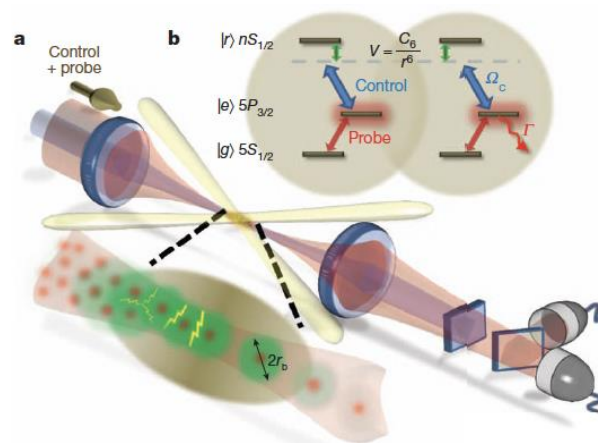
Najer *et al.* *Nature* (2019).

## Exciton polaritons



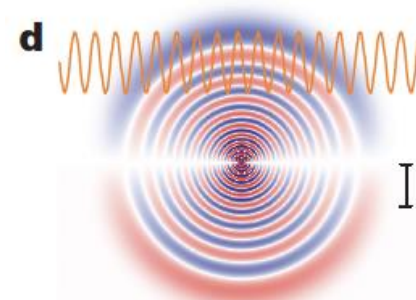
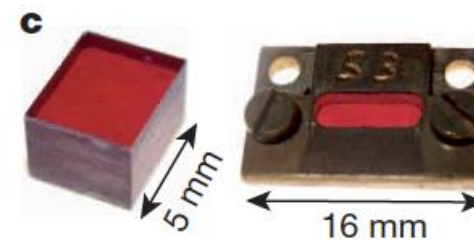
Delteil *et al.* & Matutano-Munoz *et al.* *Nature Materials* (2019).

## Cold atoms



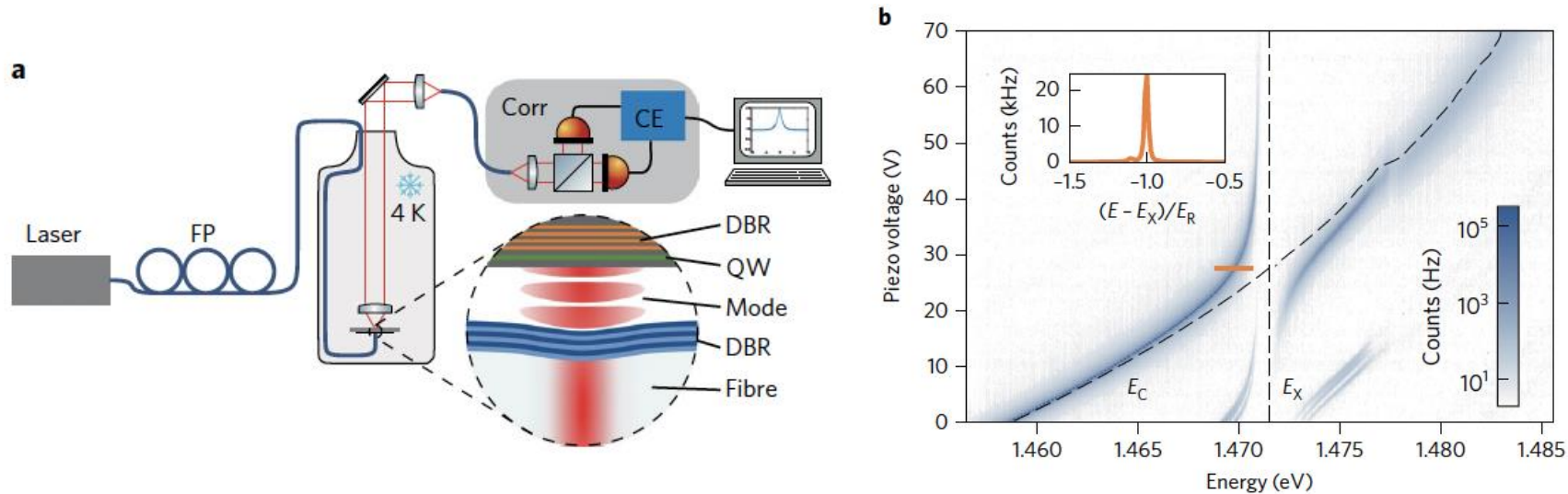
Peyronel *et al.* *Nature* (2013).

## Rydberg excitons in $\text{Cu}_2\text{O}$



Kazimierczuk *et al.* *Nature* (2014).

# Extremely strong Kerr nonlinearities from semiconductors (excitons) in microcavities



**Quantum Kerr physics in exciton-polaritons**  
Imamoglu *et al.* Physical Review Letters (1997).  
Bramati *et al.* Nature Communications (2014).  
Fink *et al.* Nature Physics (2018).  
Delteil & Fink *et al.* Nature Materials (2019).

## How close are the implementations?

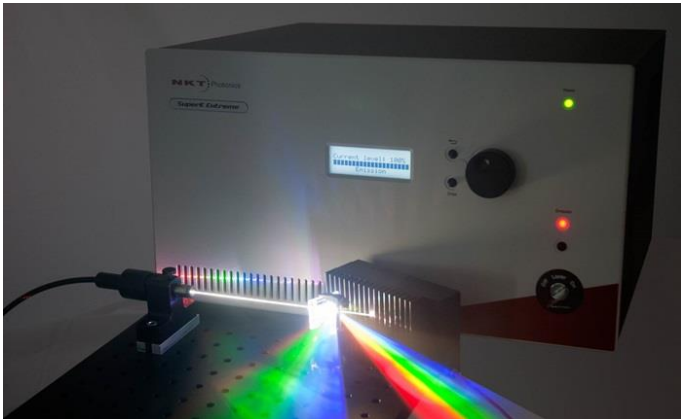
- Kerr nonlinearity within factor of 10 of loss (single-photon nonlinear regime)
- Exciton-polaritons even recently interfaced with gratings with BICs (Ardizzone *et al.* Nature (2022)).

# Why? Beyond “Gaussian” states of light

## Macroscopic nonlinear optics

Quantum state transformation

$$a_i^{\text{out}} = \alpha_i + \sum_j \mu_{ij} \delta a_j^{\text{in}} + \nu_{ij} (\delta a_j^{\text{in}})^\dagger$$



## Few-photon nonlinear optics

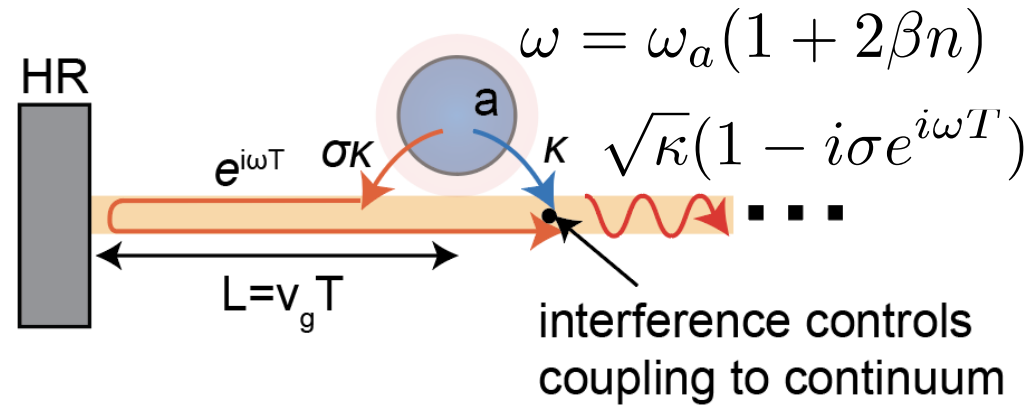
Photonic Fock states    Schrodinger “cat” states

$$|\psi\rangle \sim (a^\dagger)^n |0\rangle$$

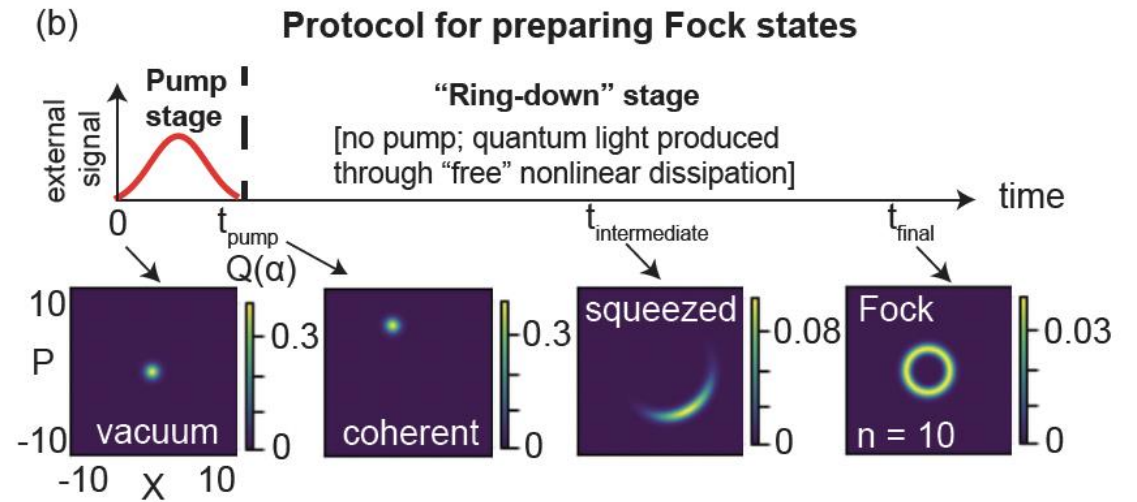
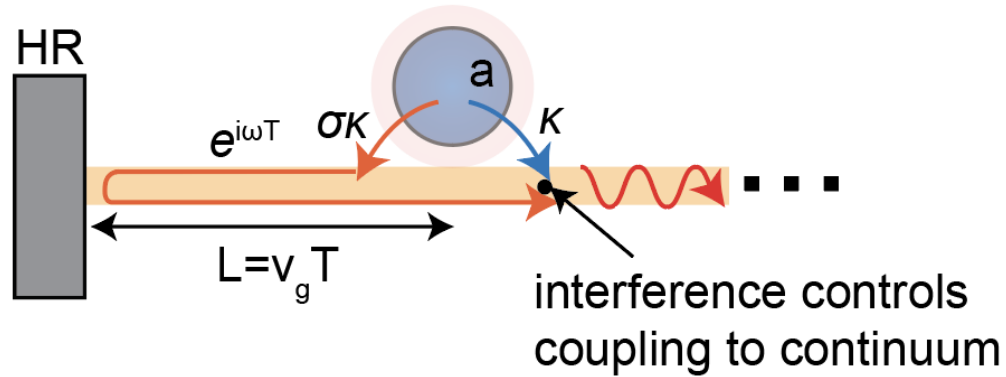
$$|\psi\rangle \sim |\alpha\rangle \pm |-\alpha\rangle$$

These and many other states have not been deterministically prepared in optics!

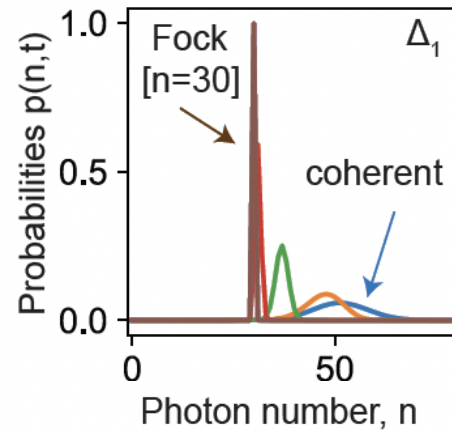
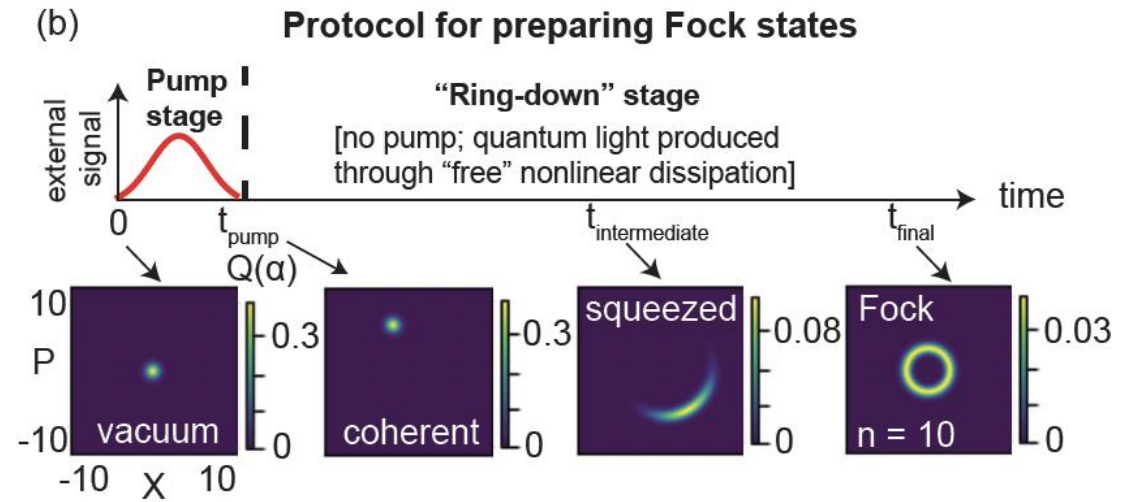
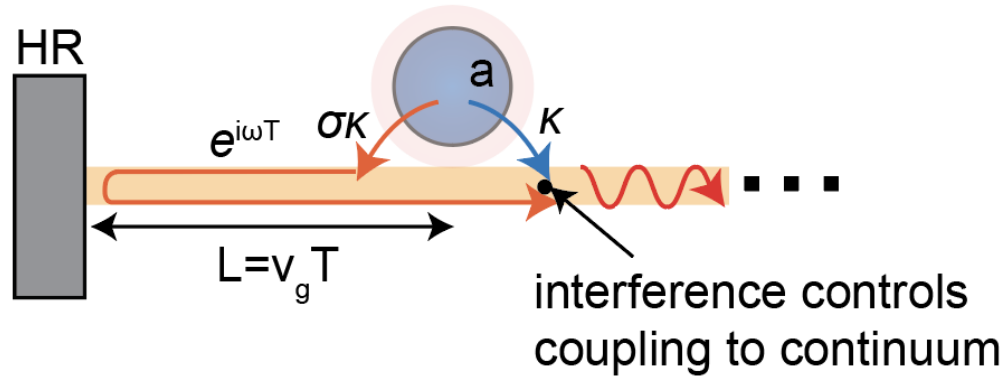
# Photonic structuring + nonlinearity: n-photon bound states



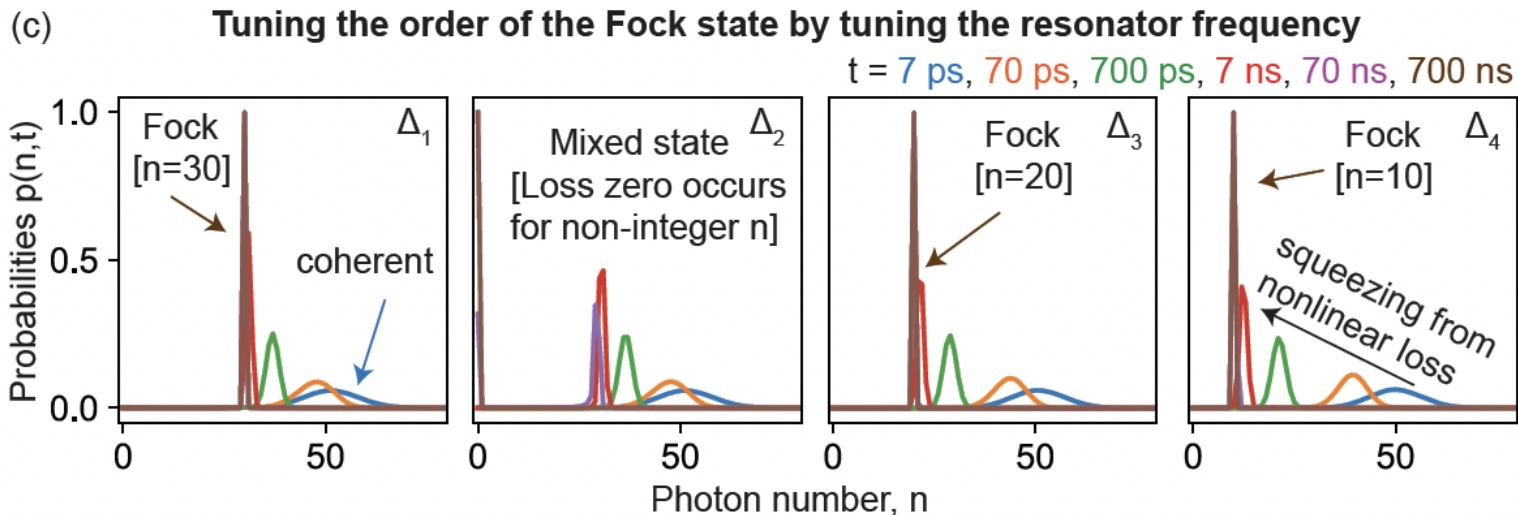
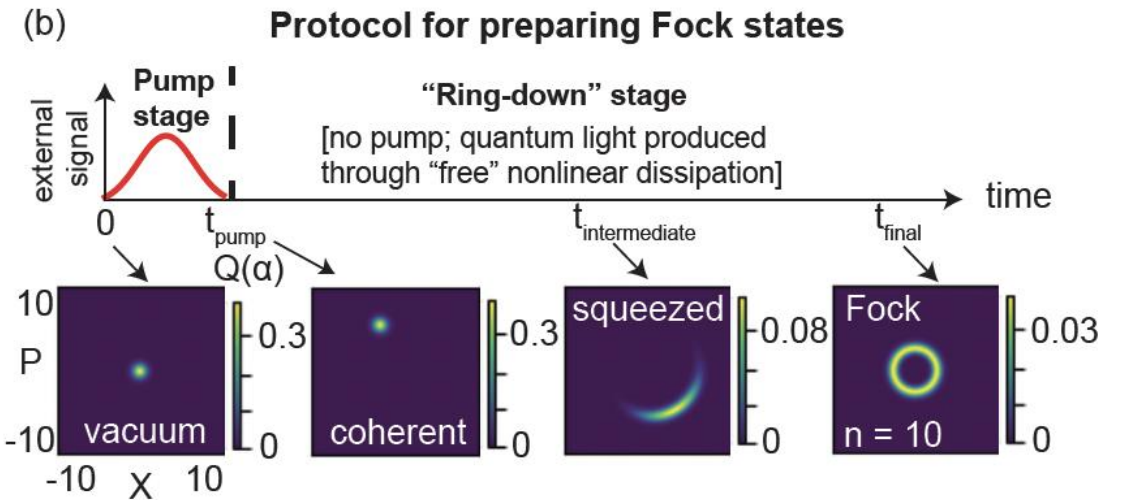
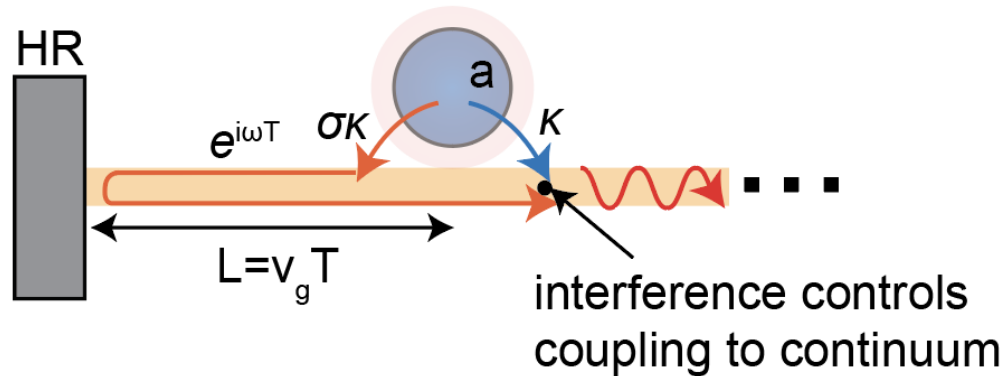
# Deterministic generation of large optical Fock states



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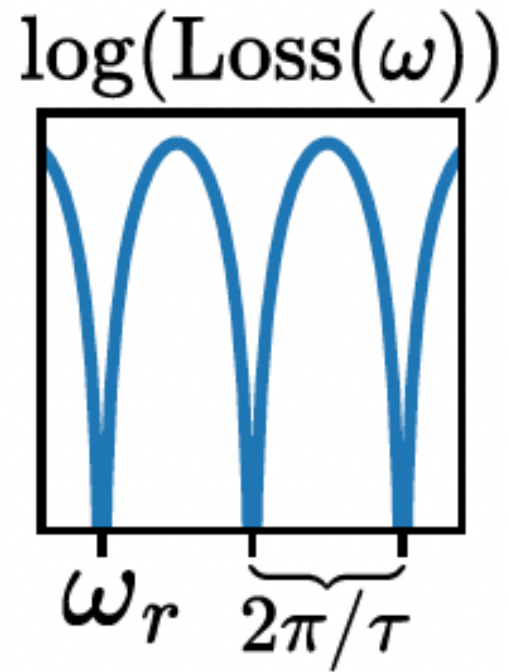


# Deterministic generation of large optical Fock states



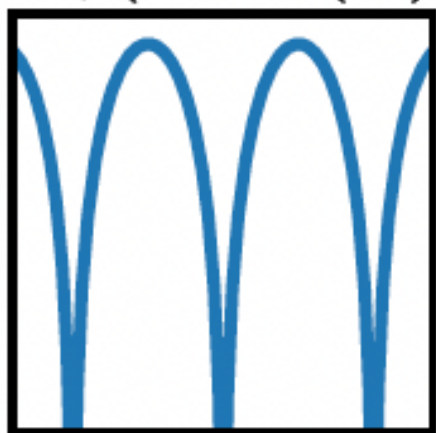
See also: Nguyen, Sloan, **Rivera et al.** PRL (2023) [low-noise lasers], Pontula\*, Sloan\*, **Rivera, et al.** arXiv:2212.07300 [THz squeezing], Sloan\*, **Rivera\* et al.** arXiv:2309.09863 [cw stabilization of Fock states]

# Controlling photonic loss: Schrodinger cat states



# Controlling photonic loss: Schrodinger cat states

$\log(\text{Loss}(\omega))$

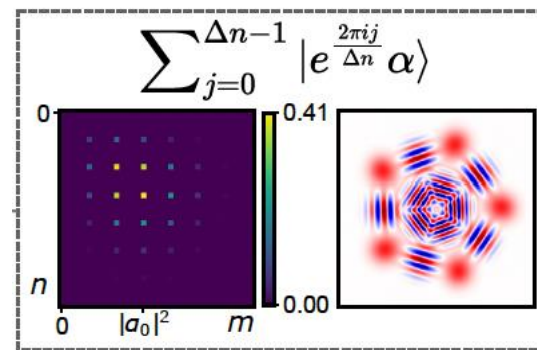
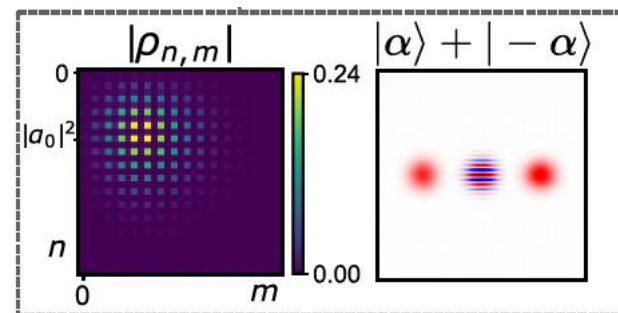


$\omega_\tau$   $2\pi/\tau$

Zero loss every 2 photons

Zero loss every 5 photons

2-legged Schrodinger cat



5-legged Schrodinger cat

**The future: quantum and nonlinear optics in new directions**

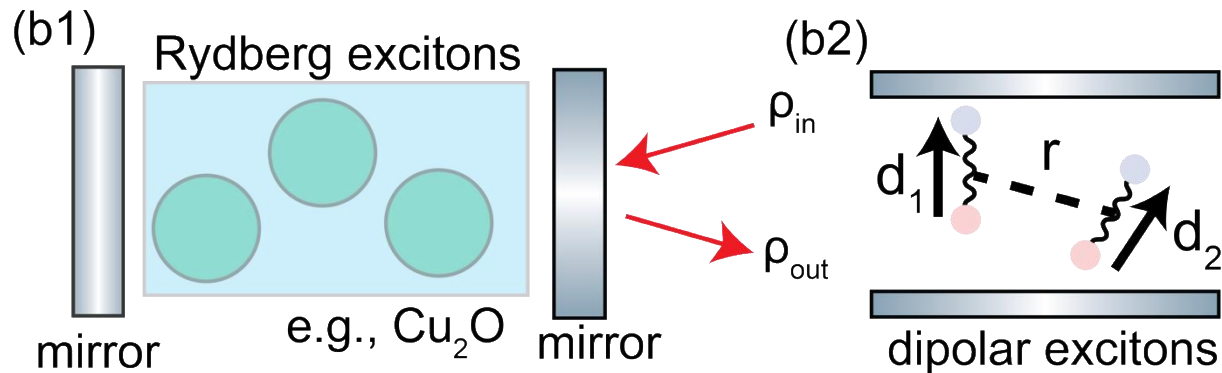
# Quantum optics in emerging material systems

**Goal:** understanding quantum light interactions with material degrees of freedom, where material degrees of freedom strongly interact. Using this to develop nonlinear optics in the "mesoscopic" regime of more than 1 or 2 photons.

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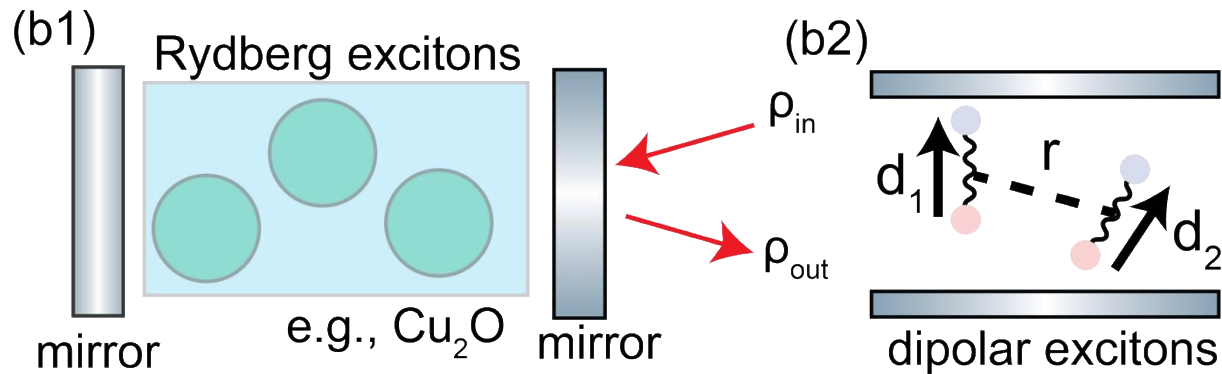
Excitonic systems: single-photon nonlinearities



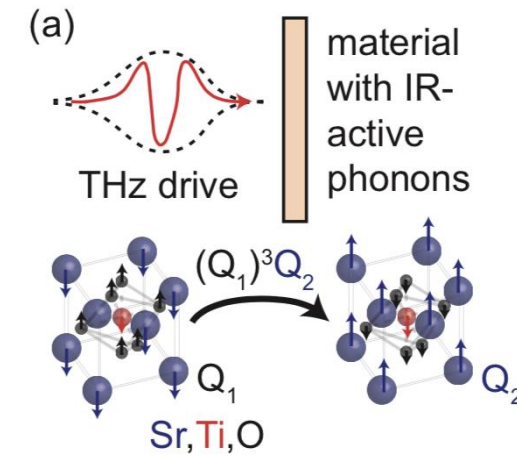
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Excitonic systems: single-photon nonlinearities



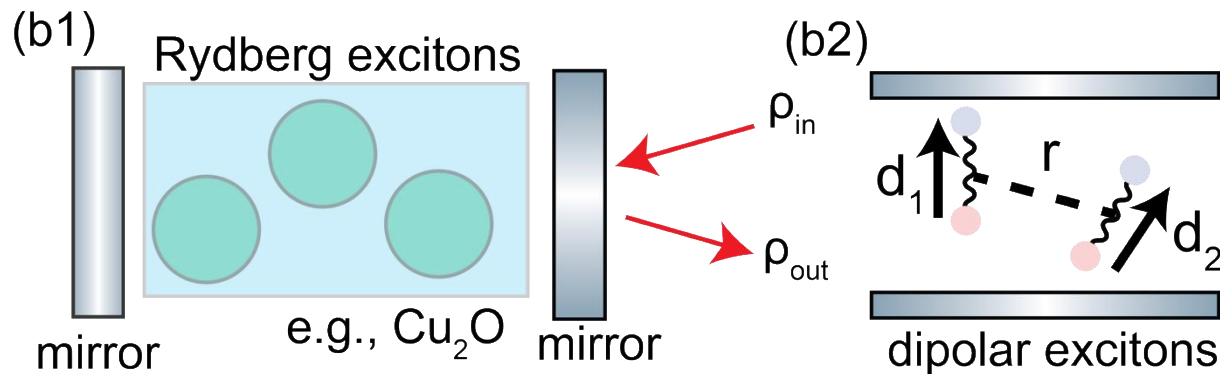
Phononic systems: quantum optics of lattice vibrations



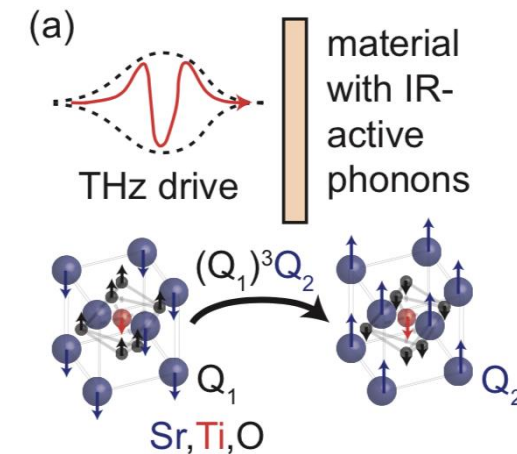
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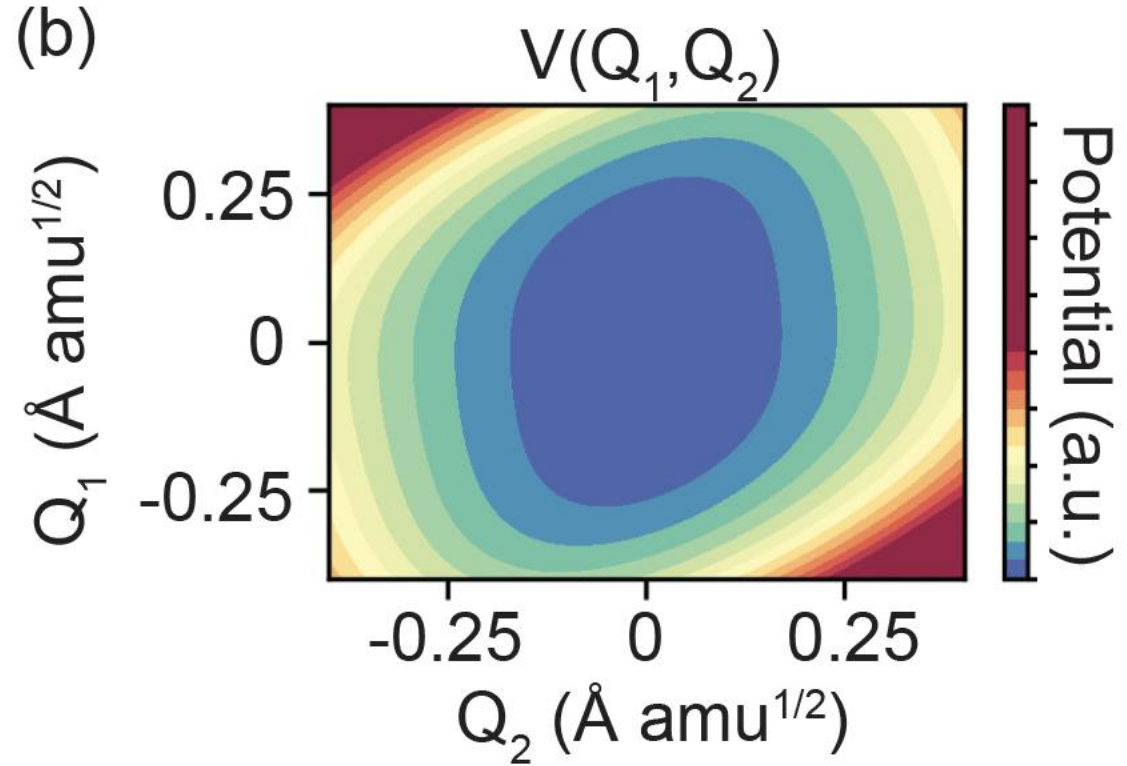
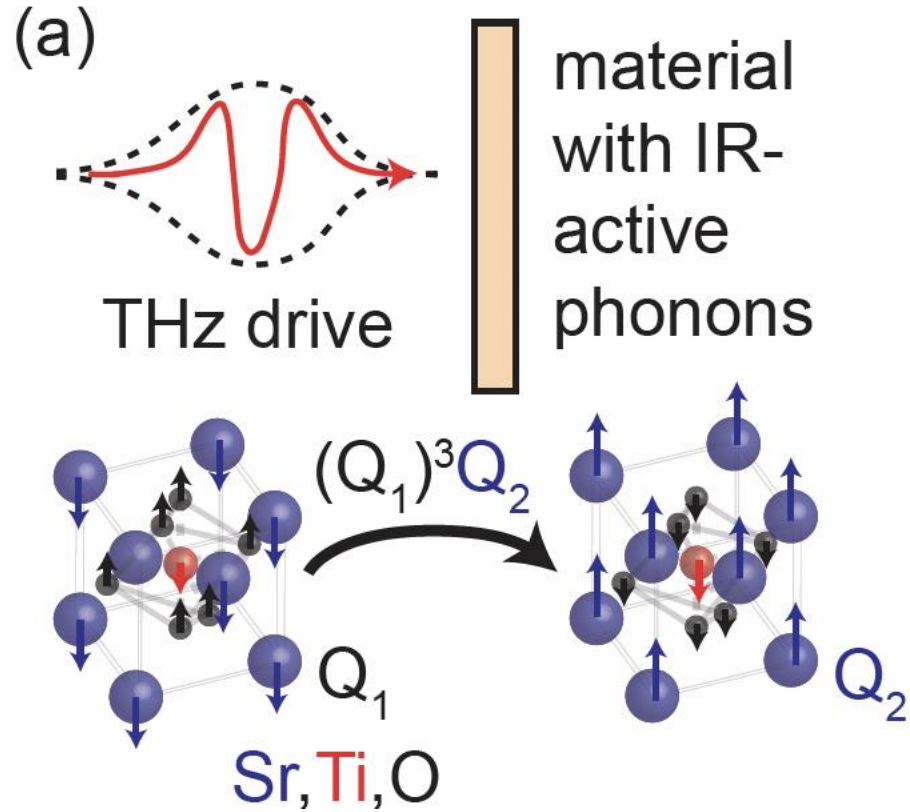


Phononic systems: quantum optics of lattice vibrations



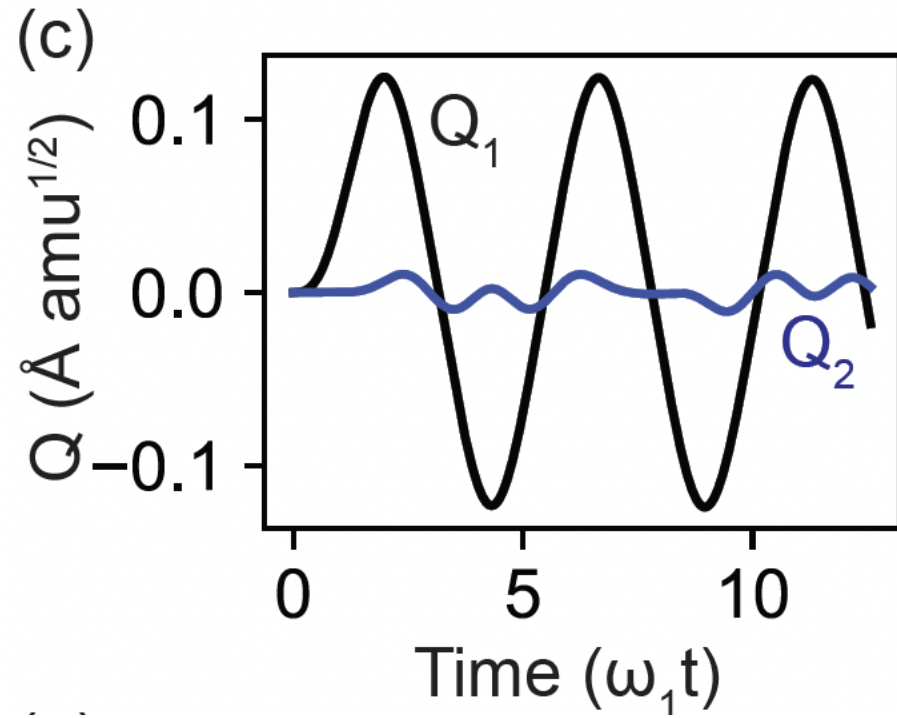
**Then what?** Excitons – understanding few-photon nonlinear response allows us to build architectures to control. Phonons – lets us develop quantum light at new frequencies (THz), and may lead to new concepts for controlling material phases

# Phonon squeezing

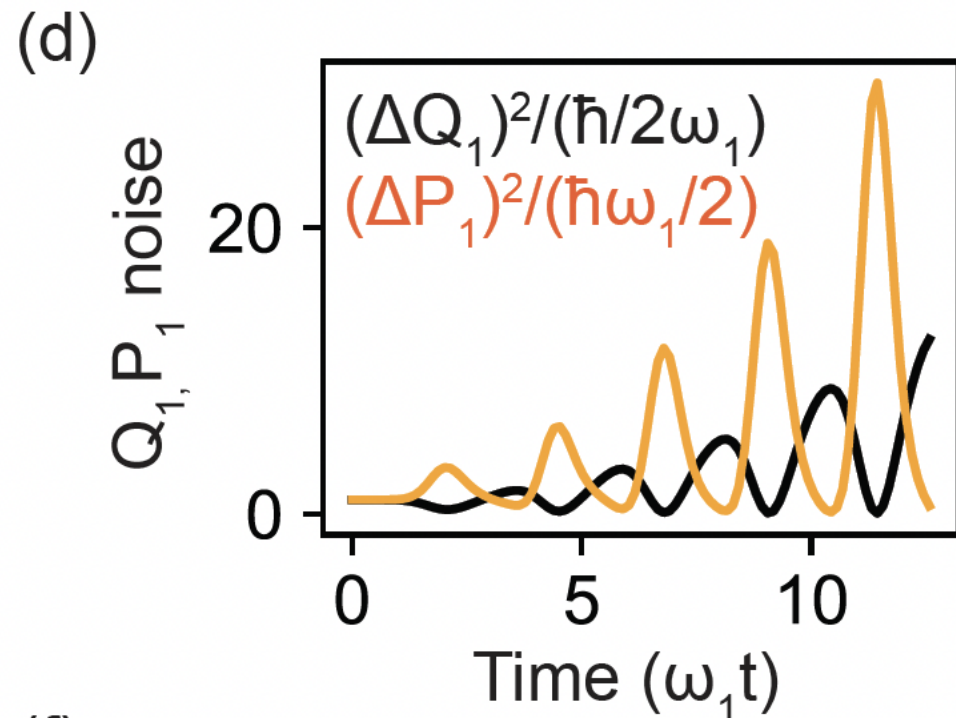
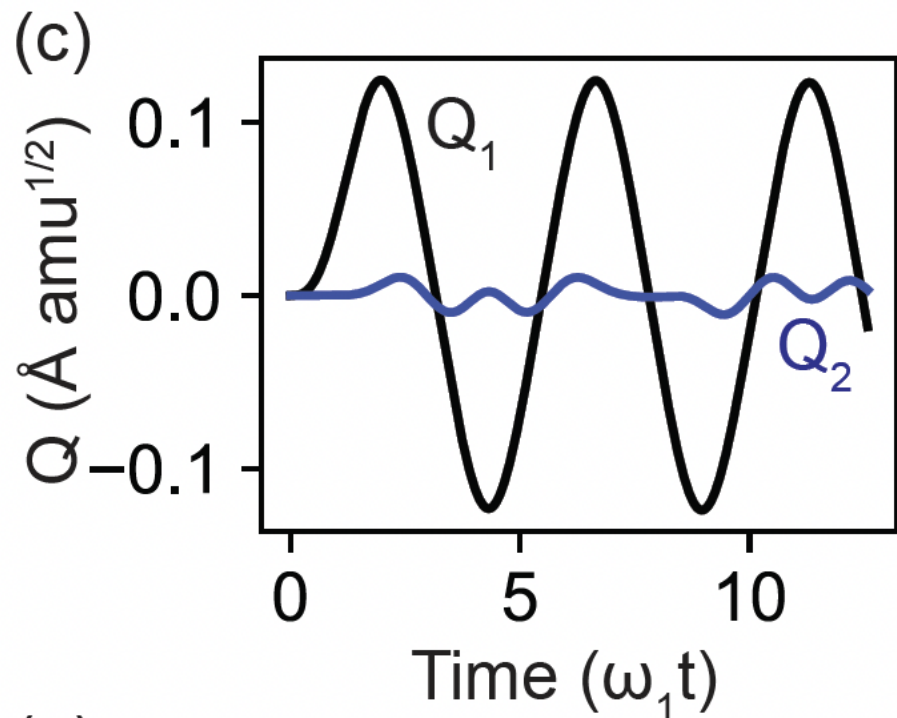


$$V = \frac{1}{2}\omega_1^2 Q_1^2 + \frac{1}{2}\omega_2^2 Q_2^2 + \frac{1}{4}\kappa_1 Q_1^4 + \frac{1}{4}\kappa_2 Q_2^4 + \chi Q_1^2 Q_2^2 + \psi_{12} Q_1^3 Q_2 + \psi_{21} Q_1 Q_2^3$$

# Phonon squeezing



# Phonon squeezing



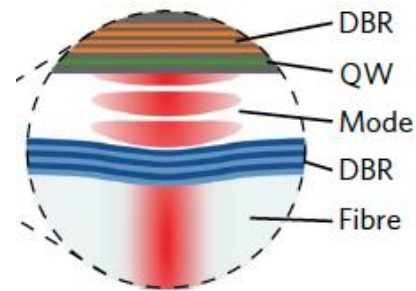
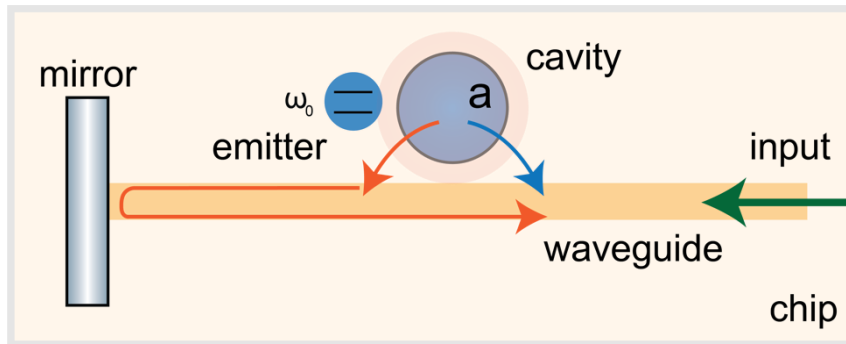
# Generating quantum light states at optical frequencies

**Goal:** exploiting few-photon nonlinear optics to *deterministically* generate non-Gaussian quantum light states (e.g., Fock, Schrodinger cat, GKP states, etc.). Addressing the gap that such schemes don't exist for multiphoton states in *optics*.

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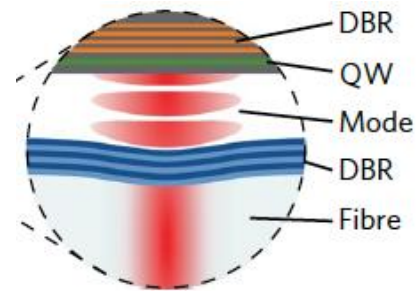
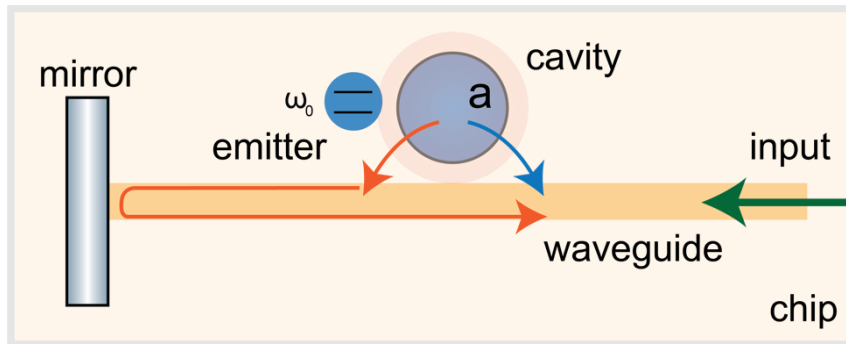
Dissipation engineering of few-photon nonlinear systems



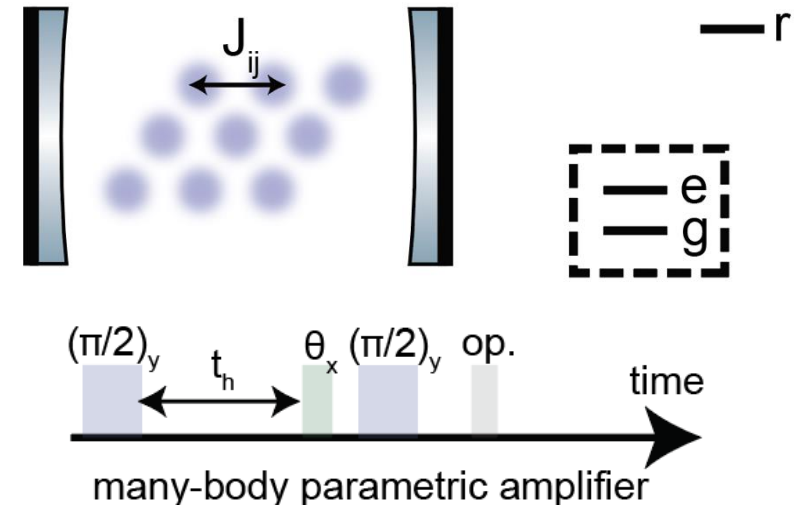
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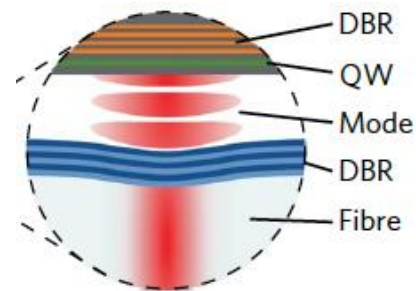
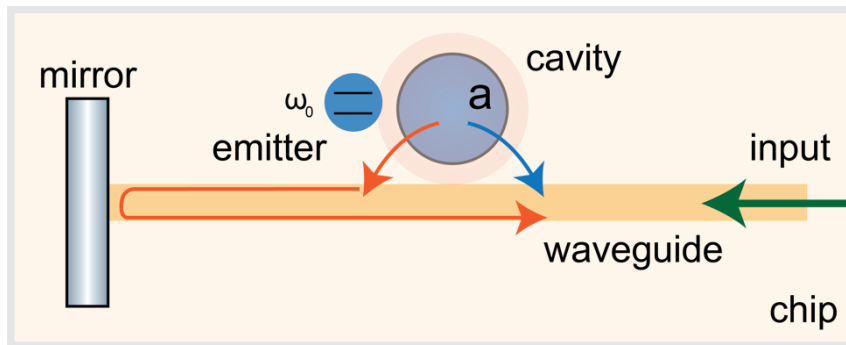
Engineering and transduction of multi-atom states



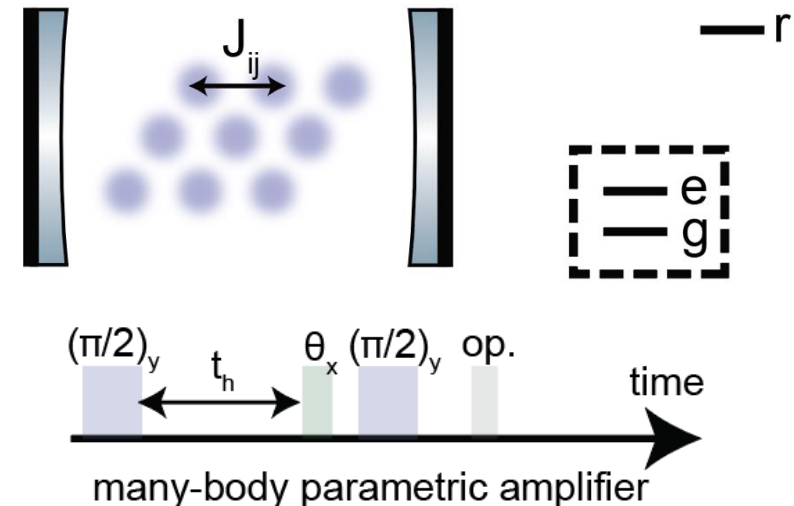
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Dissipation engineering of few-photon nonlinear systems



Engineering and transduction of multi-atom states



**Then what?** Generating certain non-Gaussian light states deterministically enables new photon-number resolving detectors, new sensing schemes, and the ability to fruitfully study driving and biasing systems with quantum light.

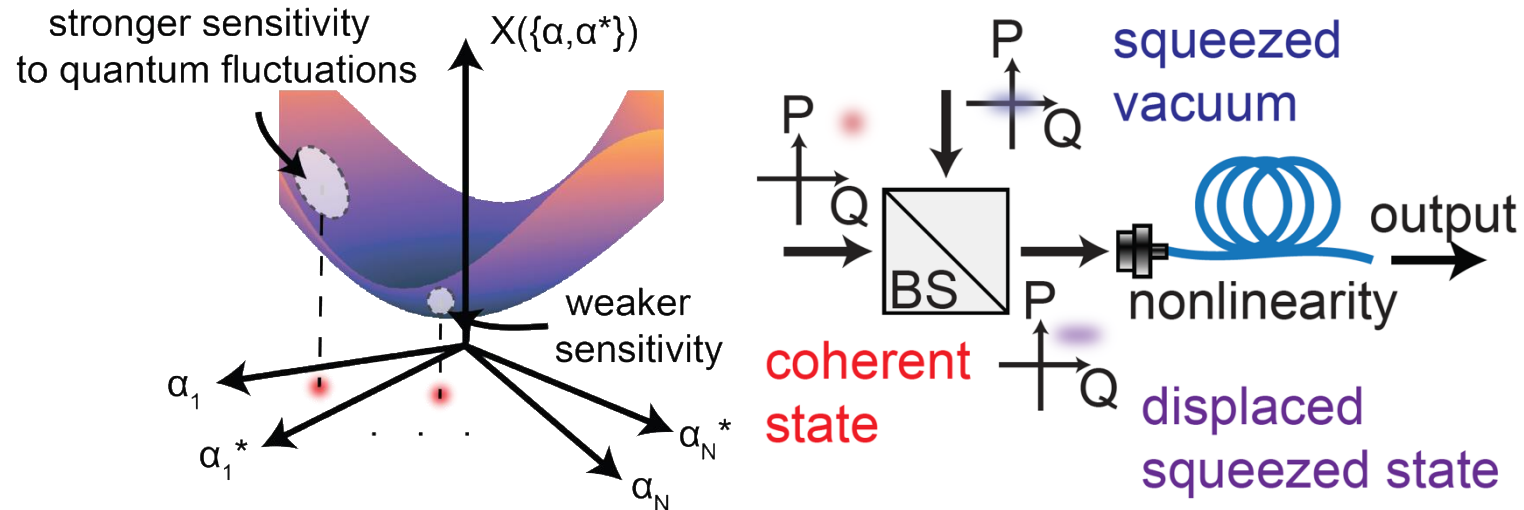
# Controlling quantum noise in multimode nonlinear systems

**Goal:** understanding quantum noise in systems where many degrees of freedom interact (in the macroscopic regime). And, using this understanding to develop foundations of a next generation of low-noise light sources in different domains.

# Controlling quantum noise in multimode nonlinear systems

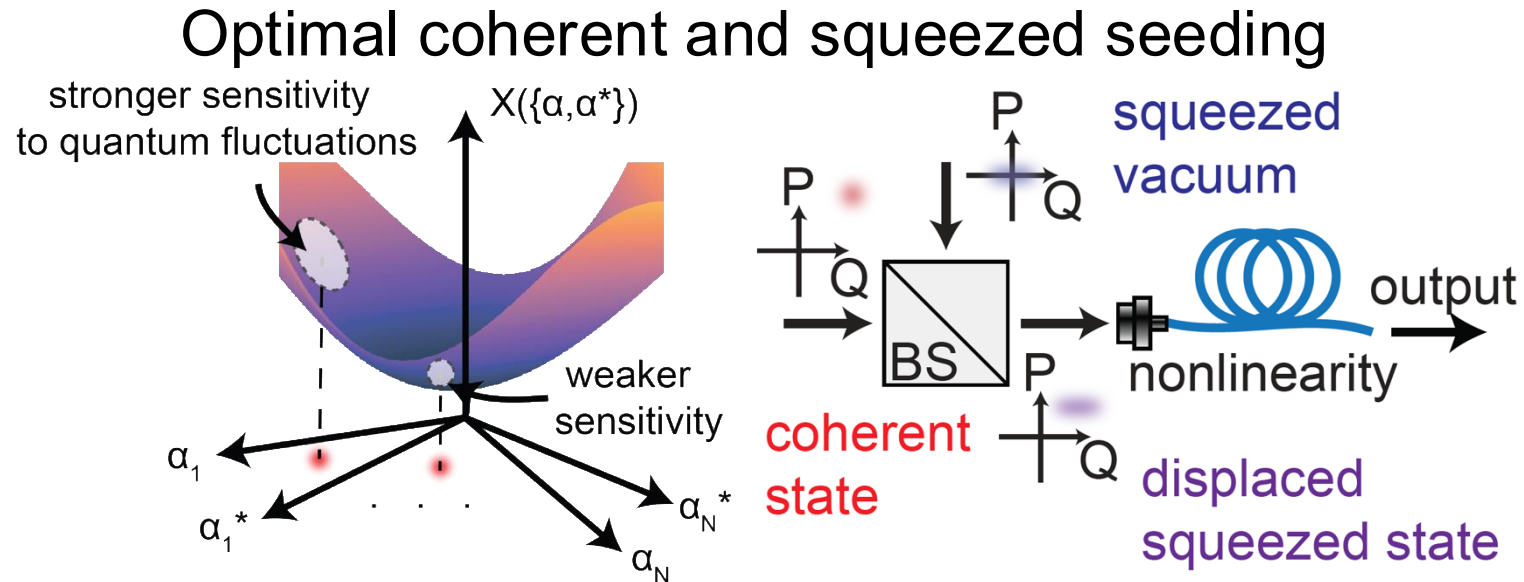
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## Optimal coherent and squeezed seeding



# Controlling quantum noise in multimode nonlinear systems

**Goal:** understanding quantum noise in systems where many degrees of freedom interact (in the macroscopic regime). And, using this understanding to develop foundations of a next generation of low-noise light sources in different domains.



**Then what?** Optimal seeding schemes for noise-limited sources like X-ray lasers, theories of quantum noise in effects like high-harmonic generation, and realizing new detectors and imaging systems based on these sources

# Acknowledgements

## Collaborators (past and ongoing)

Prof. Marin Soljačić (MIT)

Prof. John Joannopoulos (MIT)

Prof. Ido Kaminer (Technion)

Prof. Prineha Narang (Harvard, UCLA)

Prof. Dmitri Basov (Columbia)

Prof. Liang Jie Wong (NTU Singapore)

Prof. Jelena Vuckovic (Stanford)

Prof. Marko Loncar (Harvard)

Dr. Charles Roques-Carmes (MIT, Stanford)

Dr. Shiekh Zia Uddin (MIT)

Dr. Yannick Salamin (MIT)

Jamison Sloan (MIT PhD)

Ali Ghorashi (MIT PhD)

Sean Chen (MIT undergrad)

Linh Nguyen (MIT undergrad)

and many, many others!

## Funding



HARVARD  
UNIVERSITY

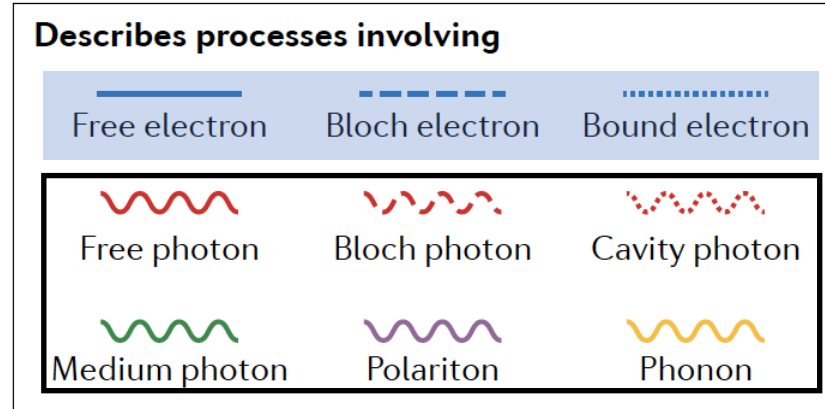
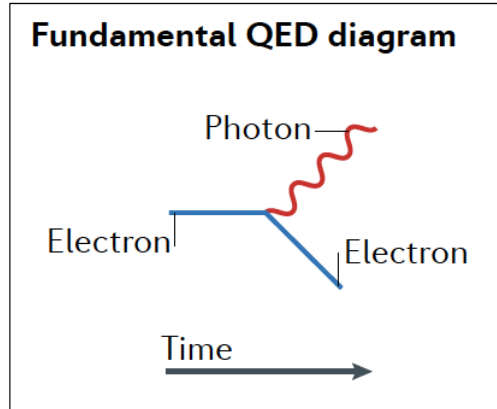


# Discussion

**Supplementary slides**

# Light-matter interactions with photonic quasiparticles

## Macroscopic quantum electrodynamics (MQED)



Free electrons or bound electrons in atoms, molecules, solids, etc.

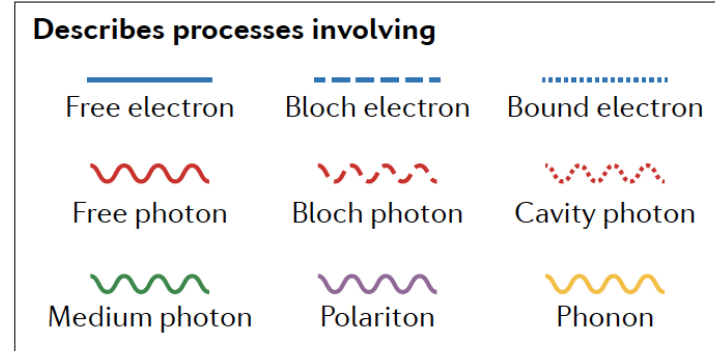
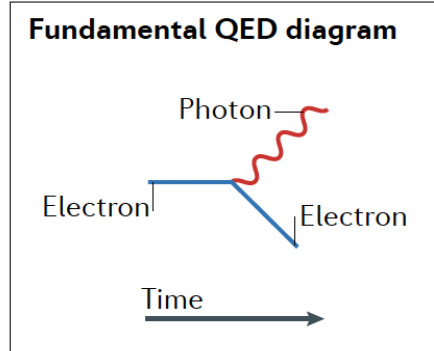
Coupling mediated by quantized EM field (e.g., vector potential)

$$\mathbf{A}(\mathbf{r}, t) = \sqrt{\frac{\hbar}{\pi\epsilon_0}} \int_{-\infty}^{\infty} d\omega e^{-i\omega t} \frac{\omega}{c^2} \int d^3r' \sqrt{\text{Im } \epsilon(\mathbf{r}', \omega)} (\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{f}(\mathbf{r}, \omega) + \text{h.c.})$$

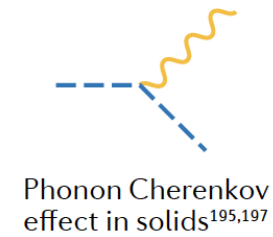
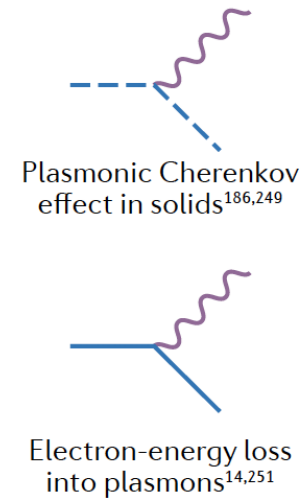
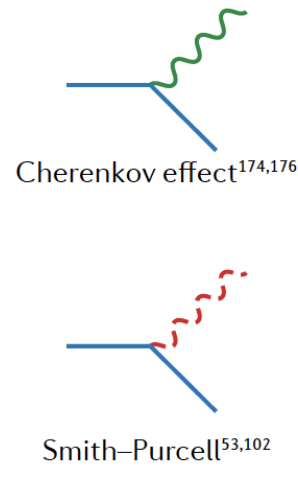
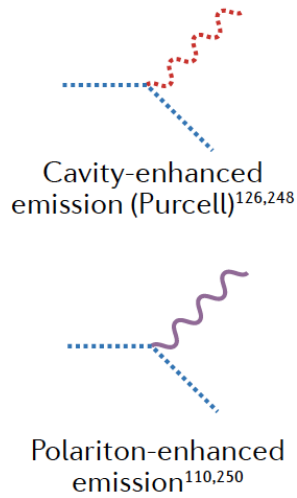
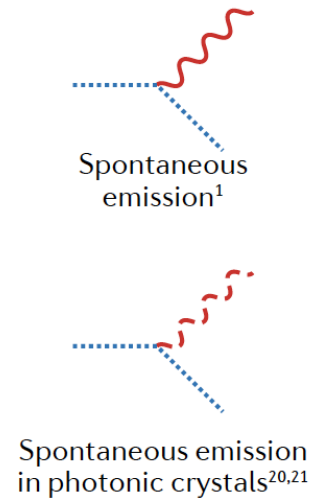
loss      field propagator:      polarization  
dispersion/modes      field operators

For applications, see: Buhmann *et al.* PRL (2008) [Casimir-Polder forces], Gonzales-Tudela *et al.* [strong coupling], Rivera *et al.* Science (2016), PNAS (2017) [higher-order effects], Goncalves *et al.* Nat. Comm. (2020) [nonlocal plasmonics], Sloan *et al.* PRL (2021) [time-dependent materials], Ben-Hayun *et al.* Sci. Adv. [free electron radiation], Svendsen *et al.* Nat. Comm. (2021), J. Chem. Theo. Comp. (2024) [interfacing with DFT].

# Elementary QED processes with photonic quasiparticles

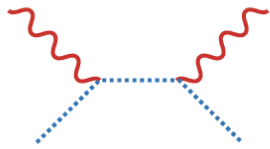


## First-order processes described by MQED

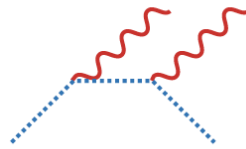


# Elementary QED processes with photonic quasiparticles

## Second-order processes described by MQED



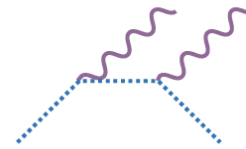
Rayleigh scattering<sup>252</sup>



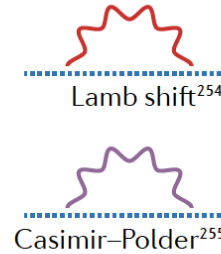
Two-photon emission<sup>122,253</sup>



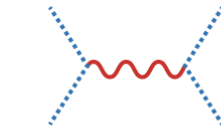
Cavity two-photon emission<sup>138</sup>



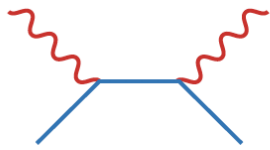
Two-polariton emission<sup>103</sup>



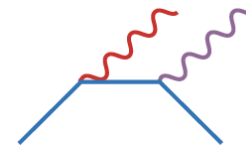
Lamb shift<sup>254</sup>



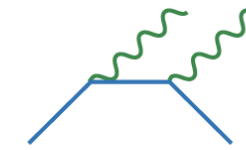
Förster resonance energy transfer<sup>256</sup>



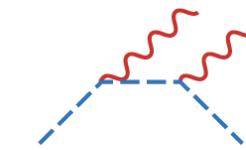
Compton scattering<sup>257</sup>



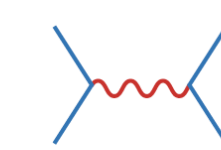
Emission of a photon and polariton<sup>122</sup>



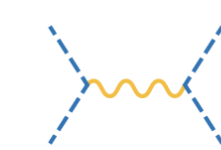
Two-photon Cherenkov effect<sup>258</sup>



Two-photon emission in solids<sup>259</sup>

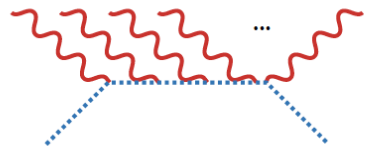


Coulomb interaction<sup>260</sup>

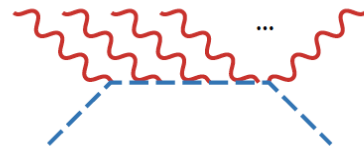


Phonon-mediated interaction<sup>261</sup>

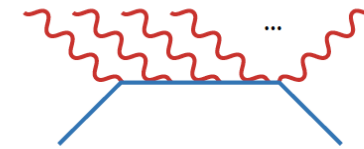
## High-order processes by MQED



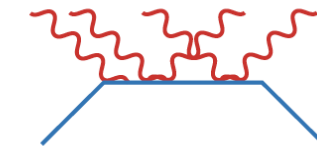
(High) harmonic generation<sup>262</sup>



(High) harmonic generation in solids<sup>262</sup>

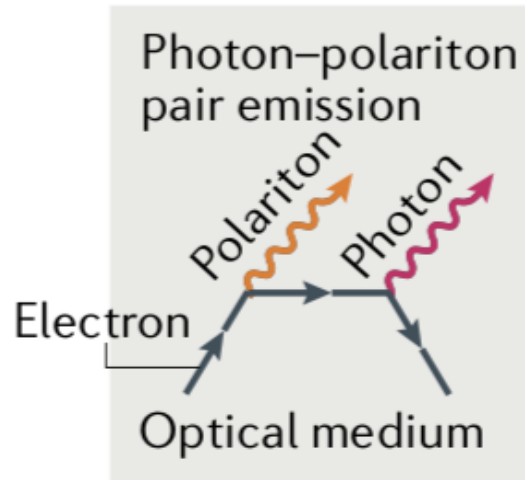


Nonlinear Compton scattering<sup>263</sup>



Photon-induced near-field electron microscopy<sup>223</sup>

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



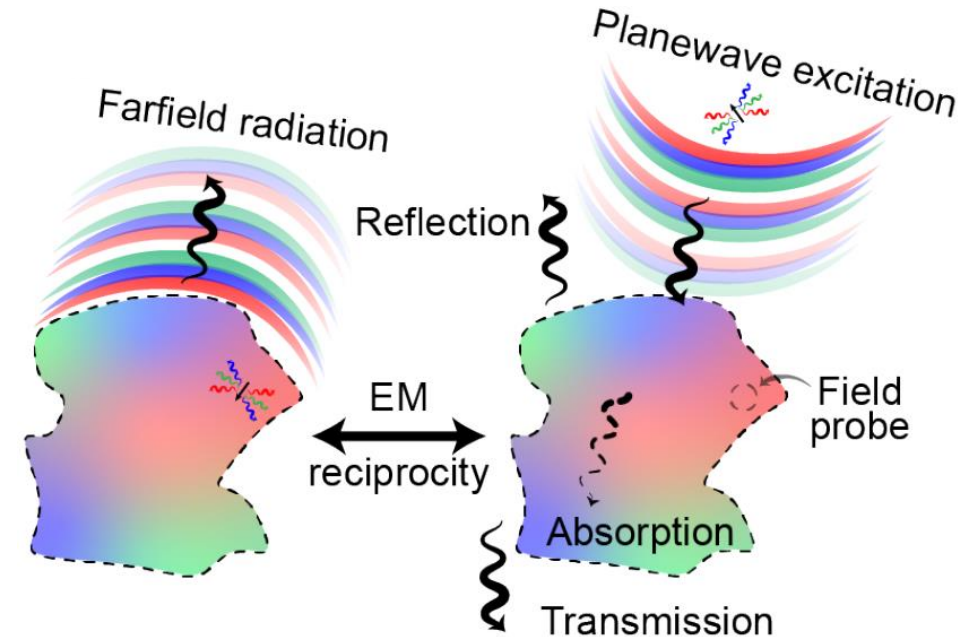
$$\omega_{\text{ph}} = \omega_{\text{pol}} \frac{\beta n(\omega_{\text{pol}}) \cos \theta_{\text{pol}} - 1}{1 - \beta \cos \theta_{\text{ph}}}$$

# Enhancing emission from fluctuating currents

Special case: uniform spatial distribution of emitters in some volume:

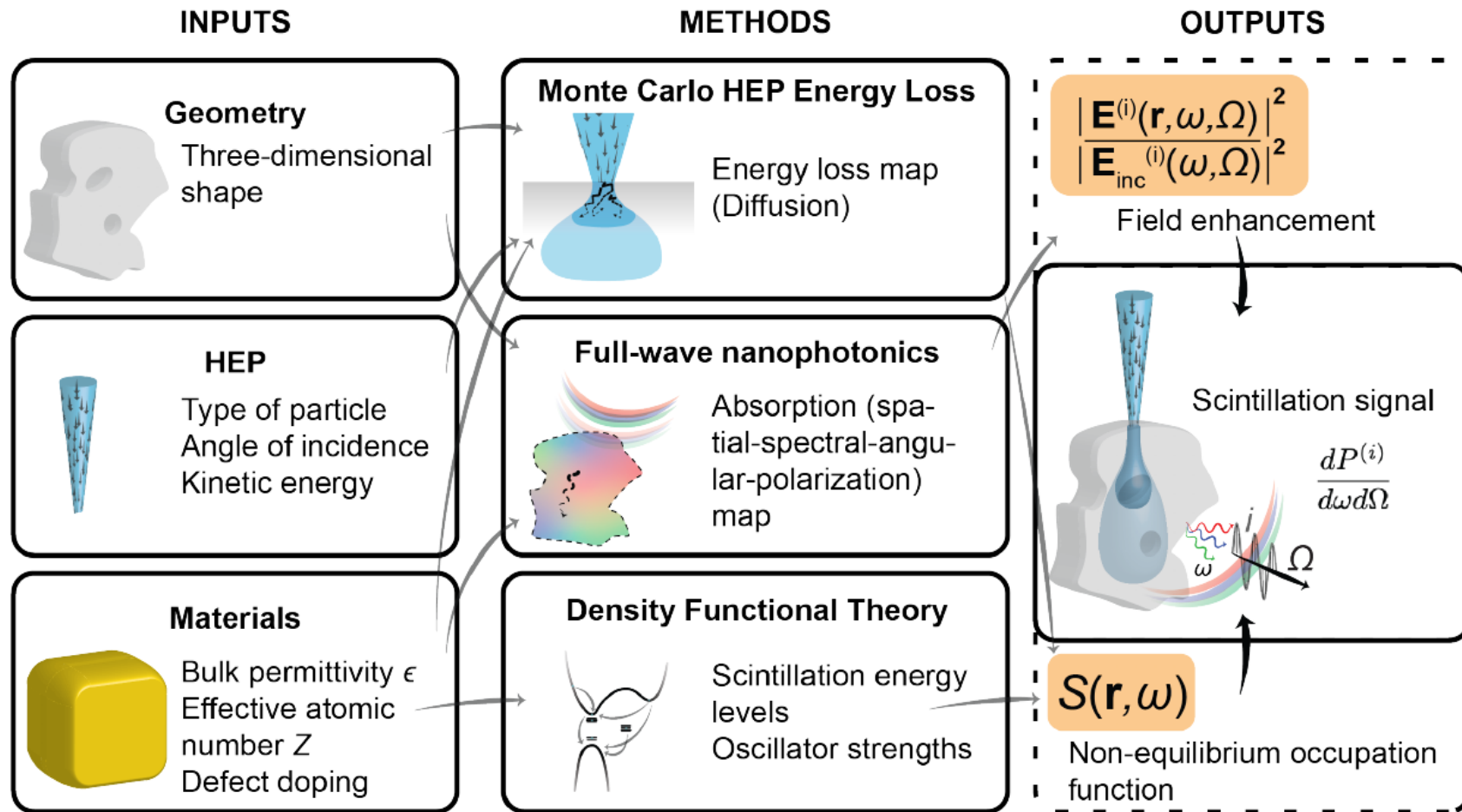
$$\frac{dP}{d\omega d\Omega} = \frac{\pi}{\epsilon_0 \omega} \times \underbrace{S(\omega)}_{\text{spectral function}} \times \underbrace{(V_{eff}(\omega)/\lambda^3)}_{\text{proportional to absorption}}$$

$$\sum_i \int d^3 r' \left| \frac{\mathbf{E}(\mathbf{r}', \mathbf{r}, i, \omega)}{E_{inc}(\mathbf{r}', \mathbf{r}, i, \omega)} \right|^2$$

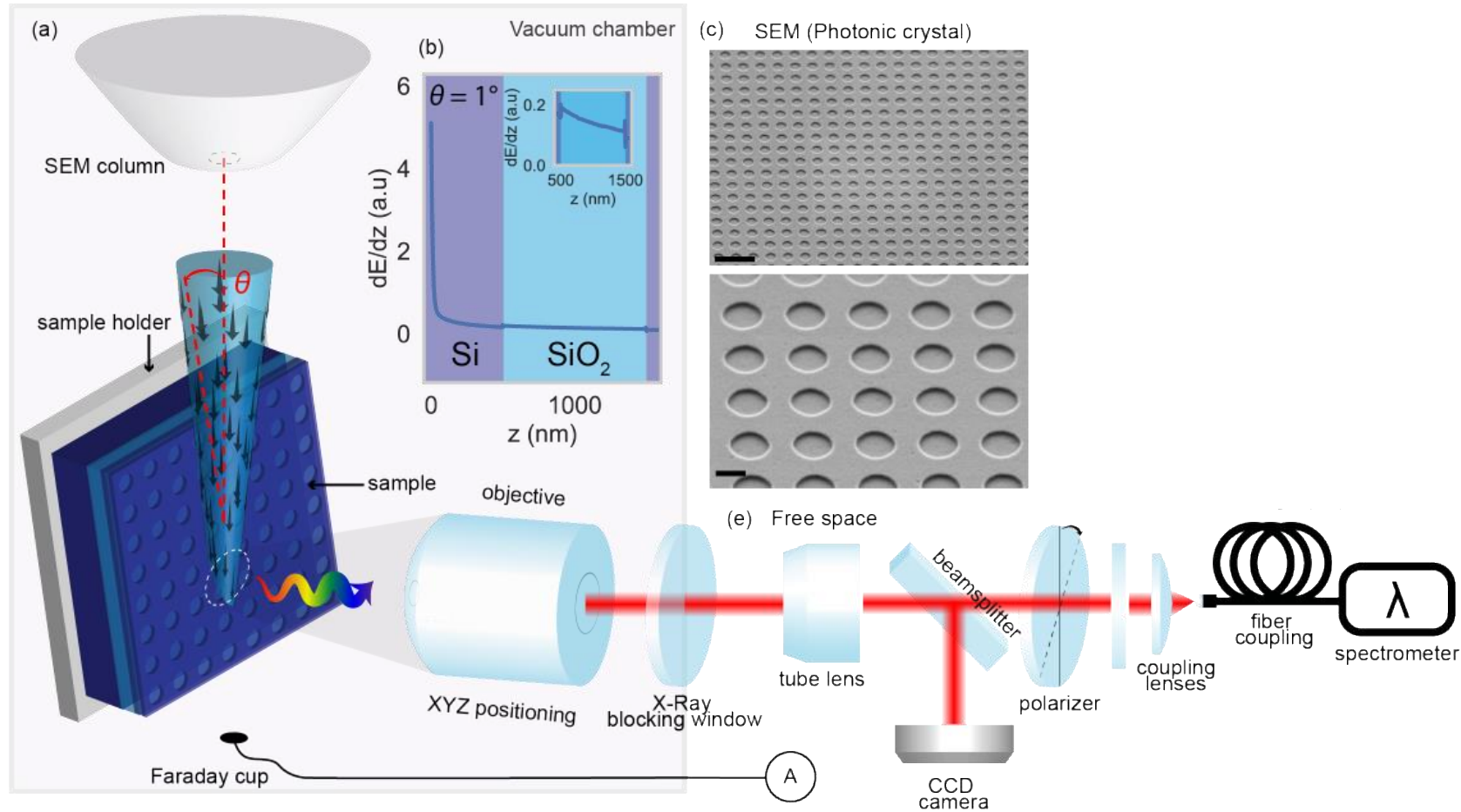


Connection to “absorbed power” means faster simulations, inverse design and optimization, and ability to borrow designs from enhanced solar cells/LEDs/thermal emission.

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

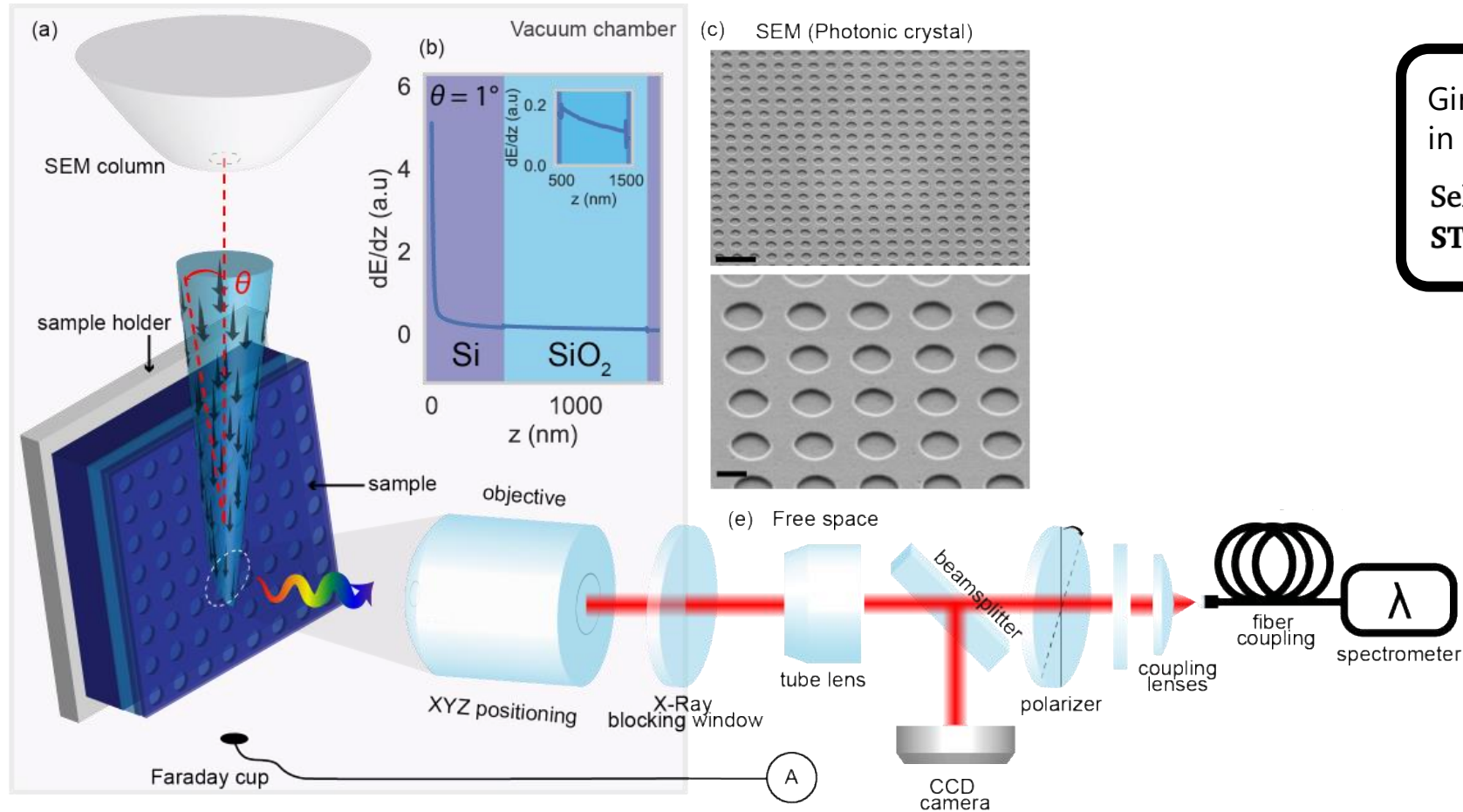


# Experiment I: enhancing scintillation induced by electrons



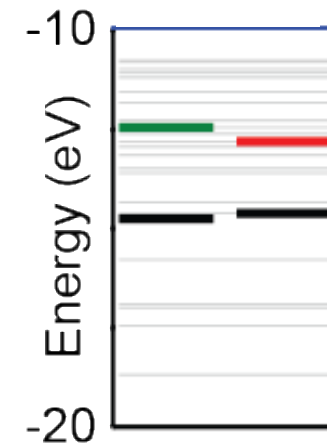
Roques-Carmes\* & Rivera\* *et al.* Science (2022).

# Experiment I: enhancing scintillation induced by electrons



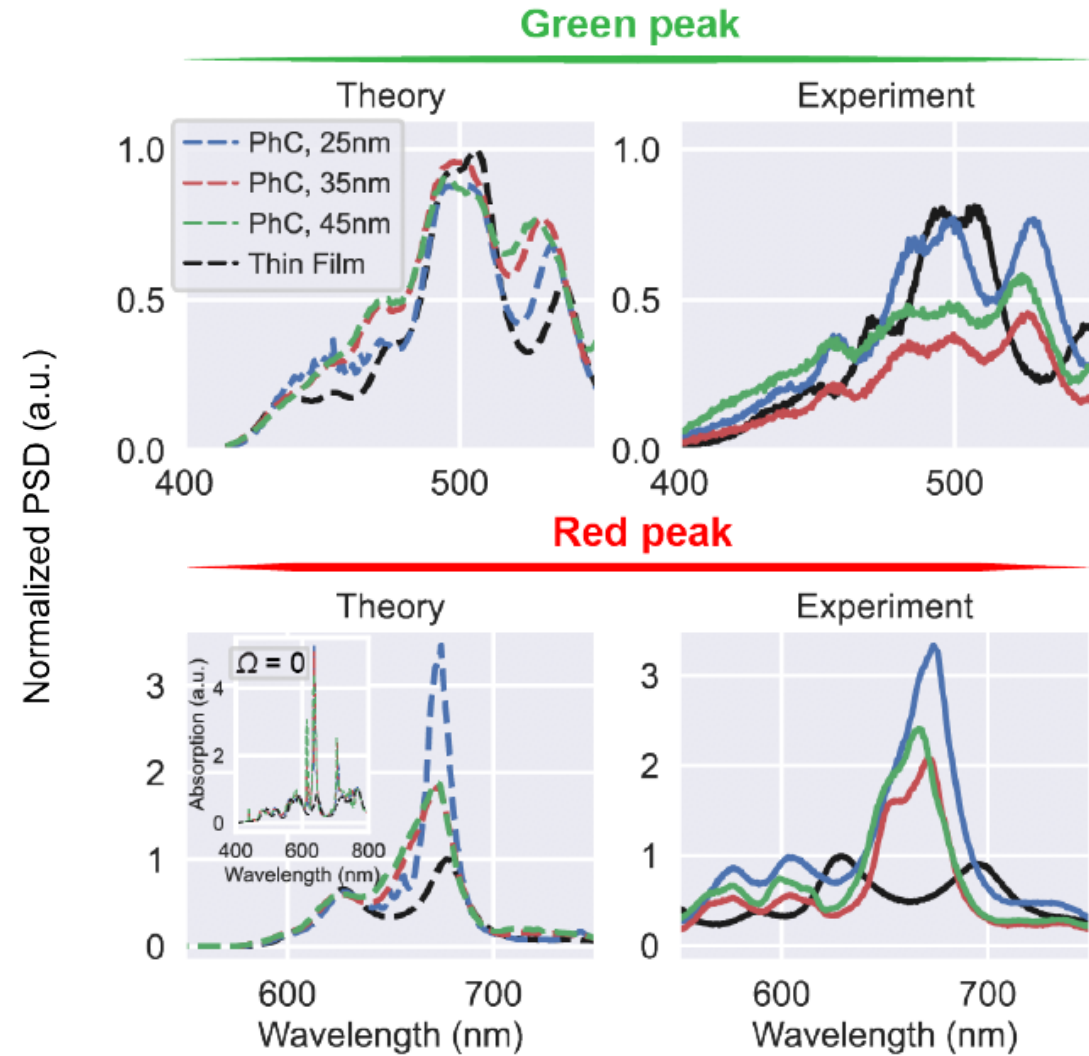
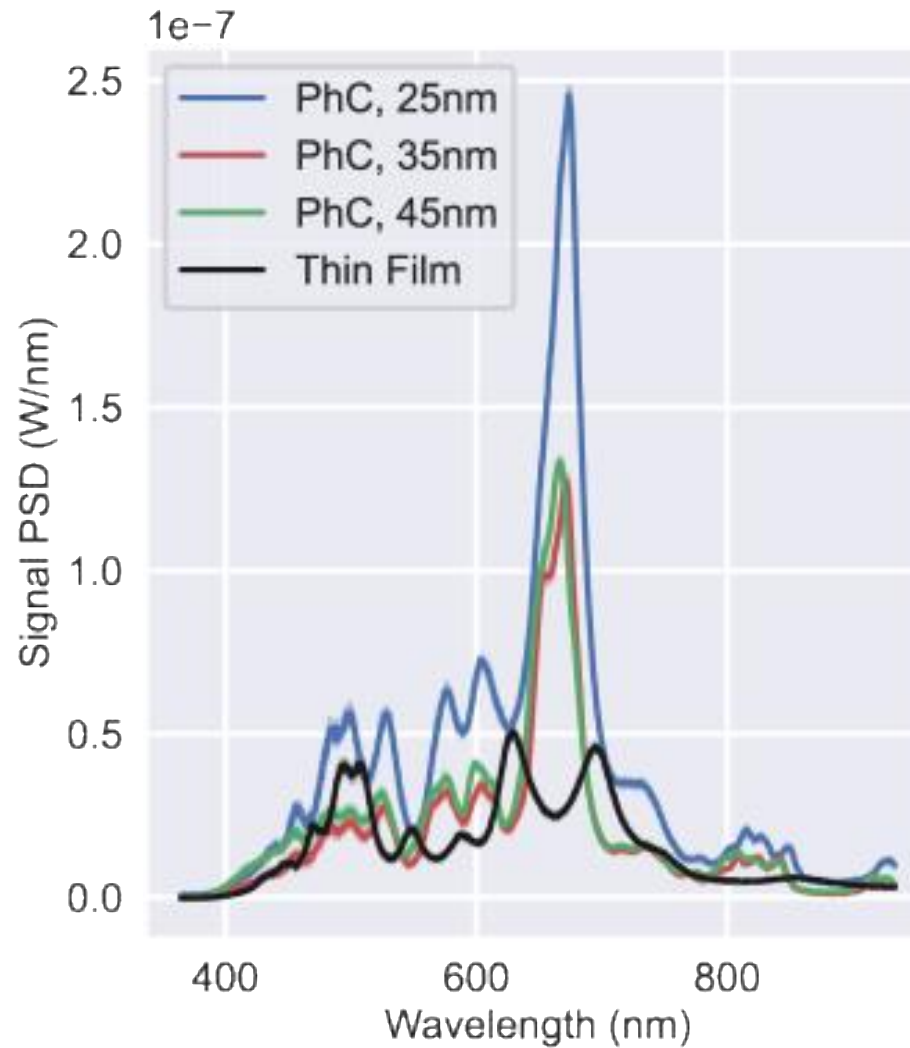
Girard et al., Reviews in Physics (2019)	<b>Optical transition (bandwidth) [eV]</b>
Self-trapped hole	2.61 (1.2)
<b>STH<sub>1</sub></b>	1,88 (0.2–0.5)

[DFT energy levels]

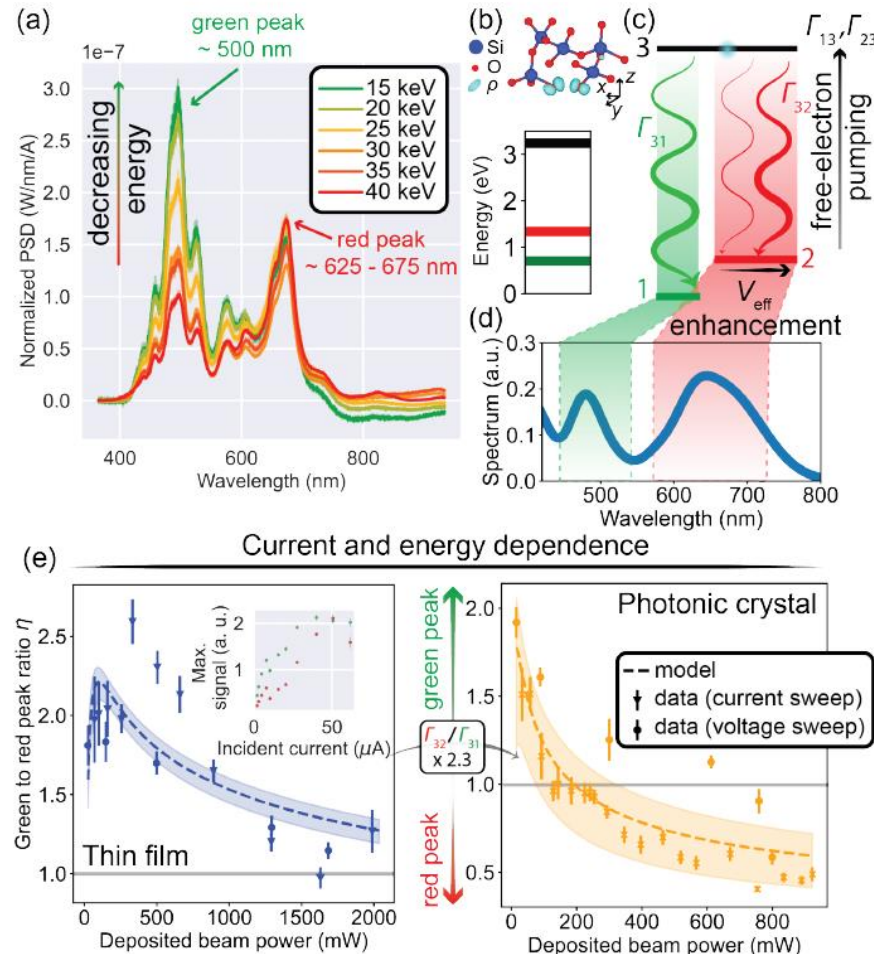


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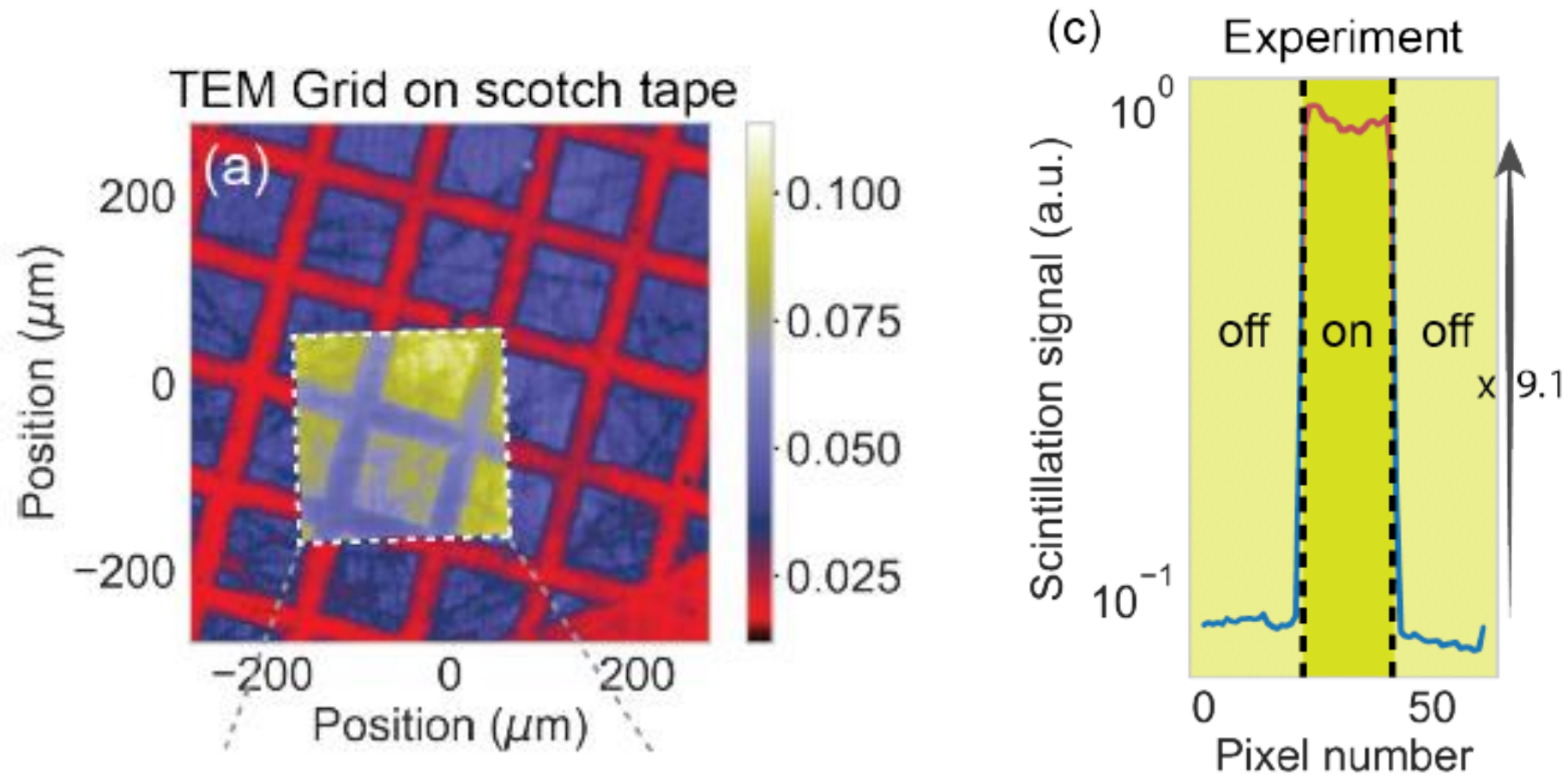
# Out-of-equilibrium kinetics of defects controlled by nanophotonics



- 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

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

# Experiment: enhancing scintillation induced by X-rays



Also did this for electron-induced scintillation: 3-6-fold enhancement.

Roques-Carmes\* and **Rivera\*** *et al.* Science (2022)

# Some immediate results of quantum sensitivity analysis

**Coherent state inputs:**  $(\Delta X)^2 = \|\nabla_{\alpha(0)} X(t)\|^2$

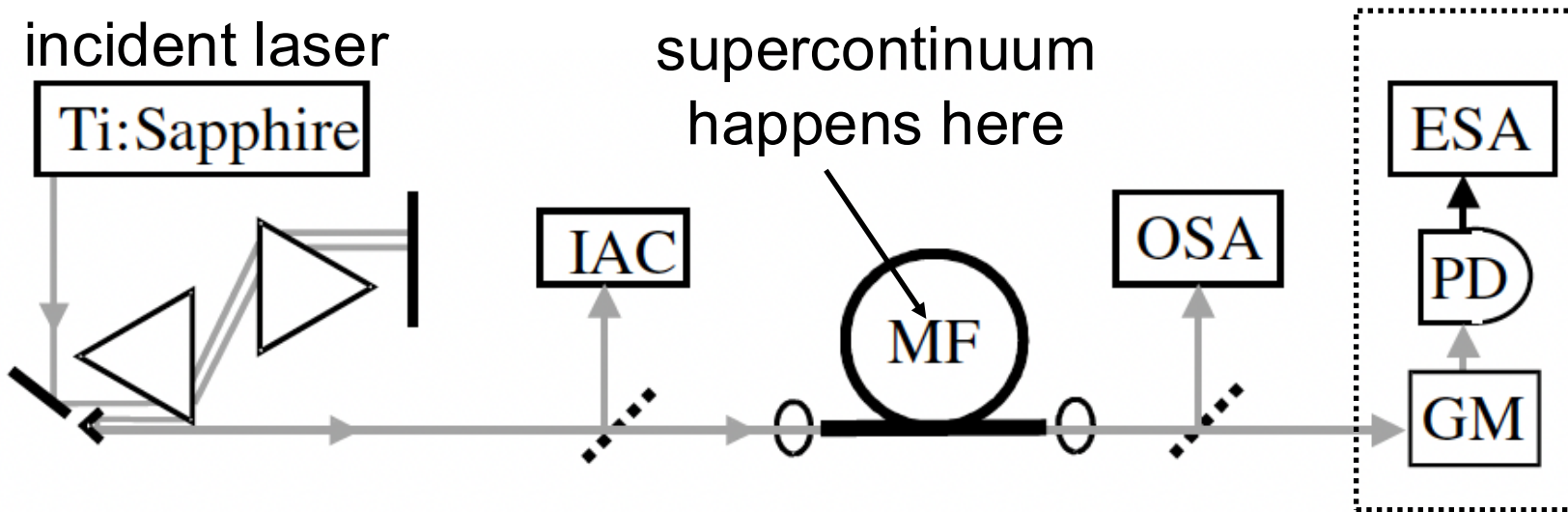
**Correlation functions:**  $\langle \delta X \delta Y \rangle = (\nabla_{\alpha(0)} X(t))^* \cdot (\nabla_{\alpha(0)} Y(t))$

**Other degrees of freedom (gain, phonons, etc.):**

$$(\Delta X)^2 = \|\nabla_{\alpha(0)} X(t)\|^2 + \|\nabla_{\beta(0)} X(t)\|^2$$

# Quantum fluctuations in supercontinuum generation

Supercontinuum is famously a noisy process, and has a fundamental quantum origin: amplification of incident vacuum noise.



Corwin *et al.*  
*PRL* (2003).

Nearly 1-10% pulse-to-pulse fluctuation from inputs with  $<0.01\%$ . Individual wavelengths are not in coherent states, even when initial light is!

# The classical description of supercontinuum generation

incident fs pulse:  $A(z,t)$

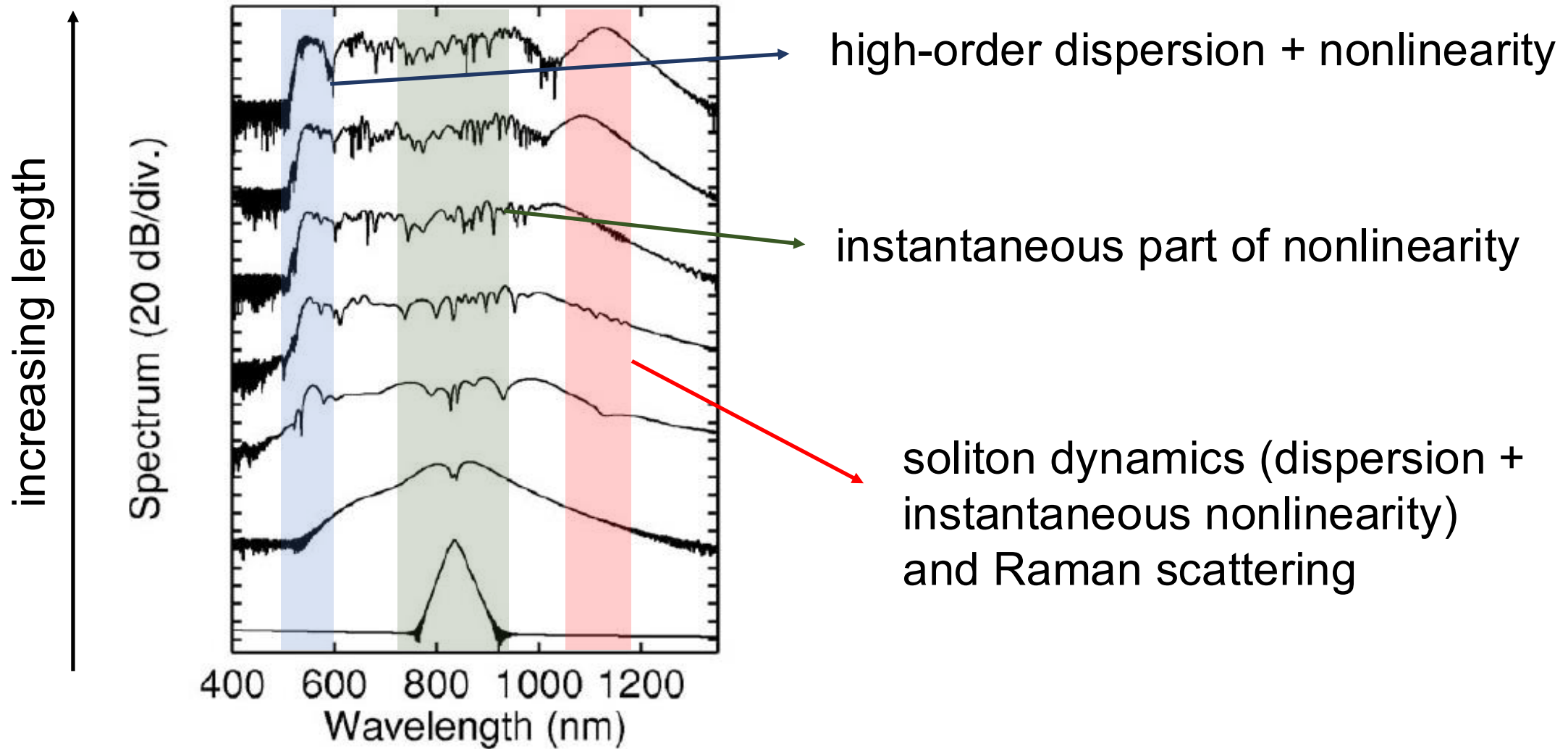


**Pulse evolution: the generalized nonlinear Schrodinger equation**

$$\partial_z A(z, t) = \underbrace{-\frac{\alpha}{2} A(z, t)}_{\text{loss}} + \underbrace{\sum_{k=2}^{\infty} \frac{i^{k+1} \beta_k}{k!} \partial_t^k A(z, t)}_{\text{dispersion: } k = k(\omega)} + \underbrace{i\gamma A(z, t) \int dt' R(t') |A(z, t - t')|^2}_{\text{Nonlinearity: instantaneous (electronic) + delayed (phonons)}}$$

This equation (+ variants) has been studied by many authors for a long time (50+ years)!

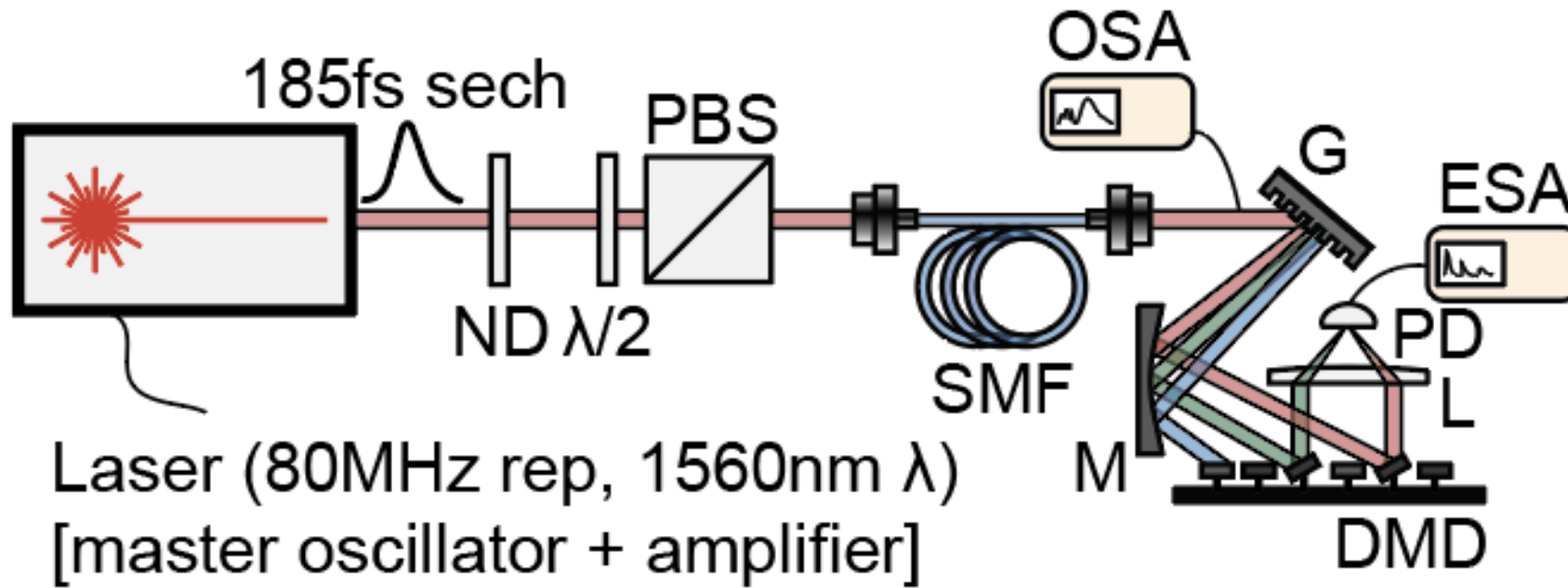
# The physical processes leading to supercontinuum generation



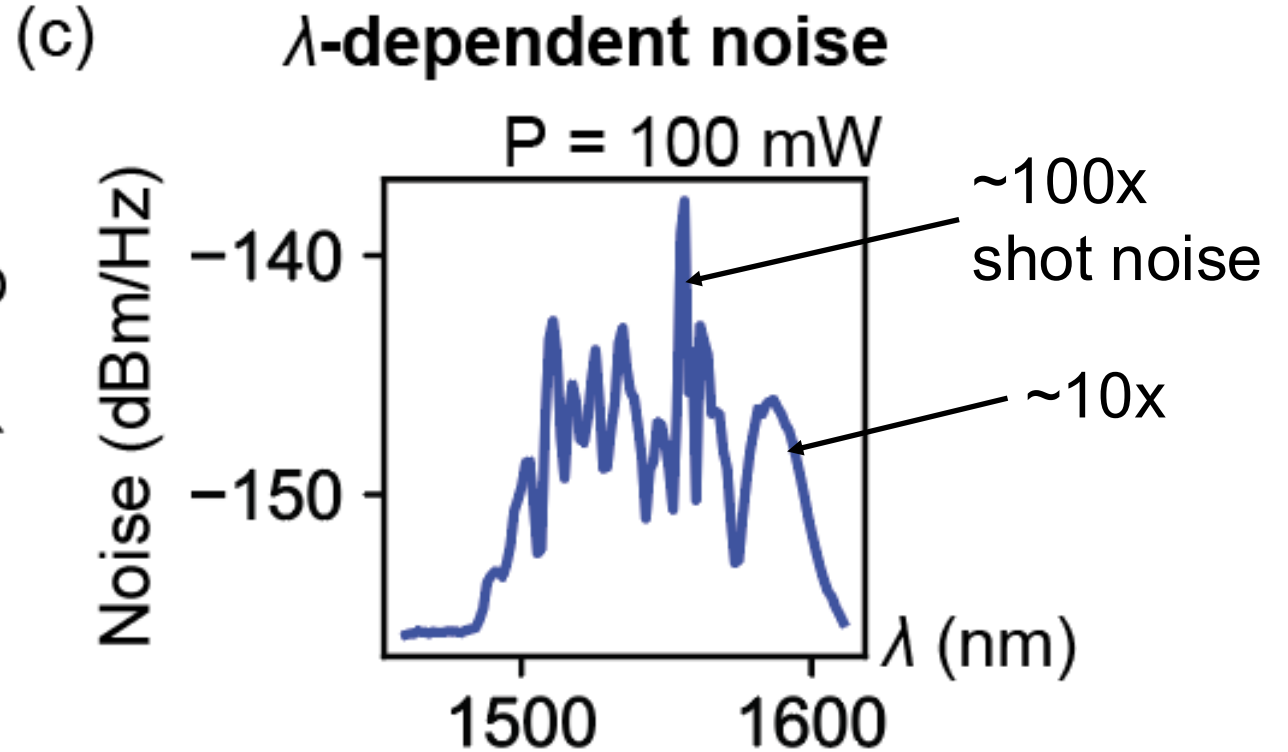
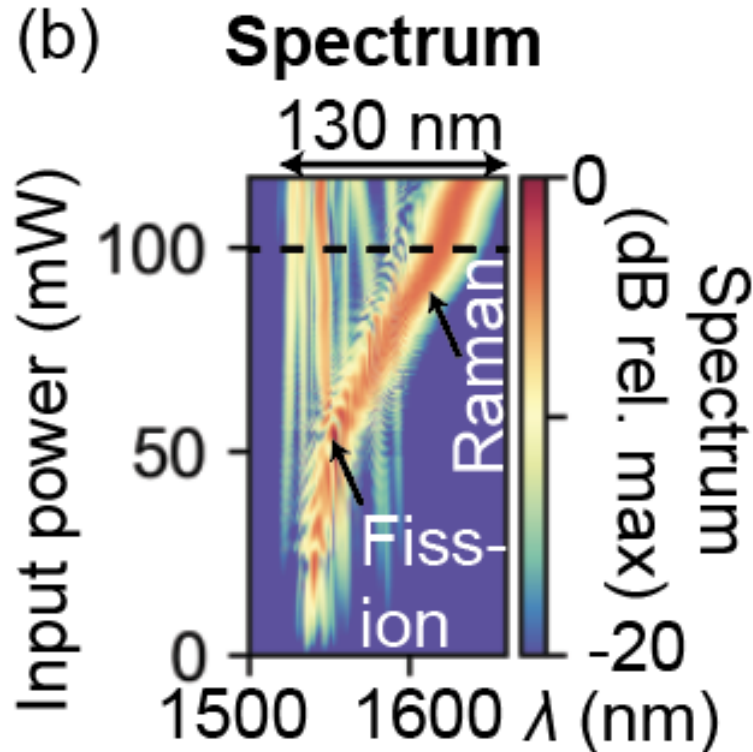
Dudley *et al.* *Rev. Mod. Phys.* (2006).

# A new low noise regime in supercontinuum generation

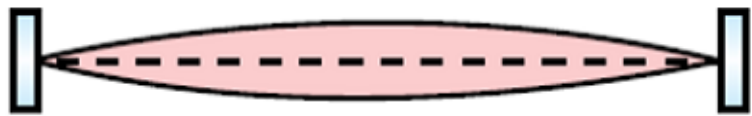
(a) Experimental schematic



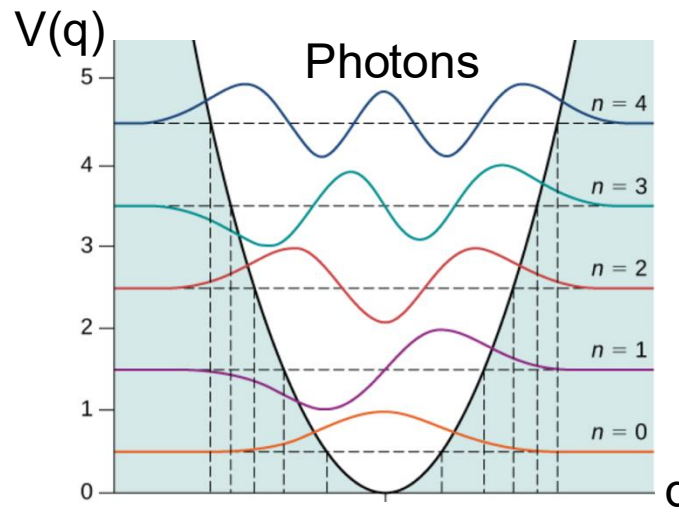
# A new low noise regime in supercontinuum generation



# Fock states are fundamental, broadly applicable, and difficult to create



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



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

- In optics,  $n \geq 2$  is difficult to produce
- Proposed techniques are generally non-deterministic

## Applied

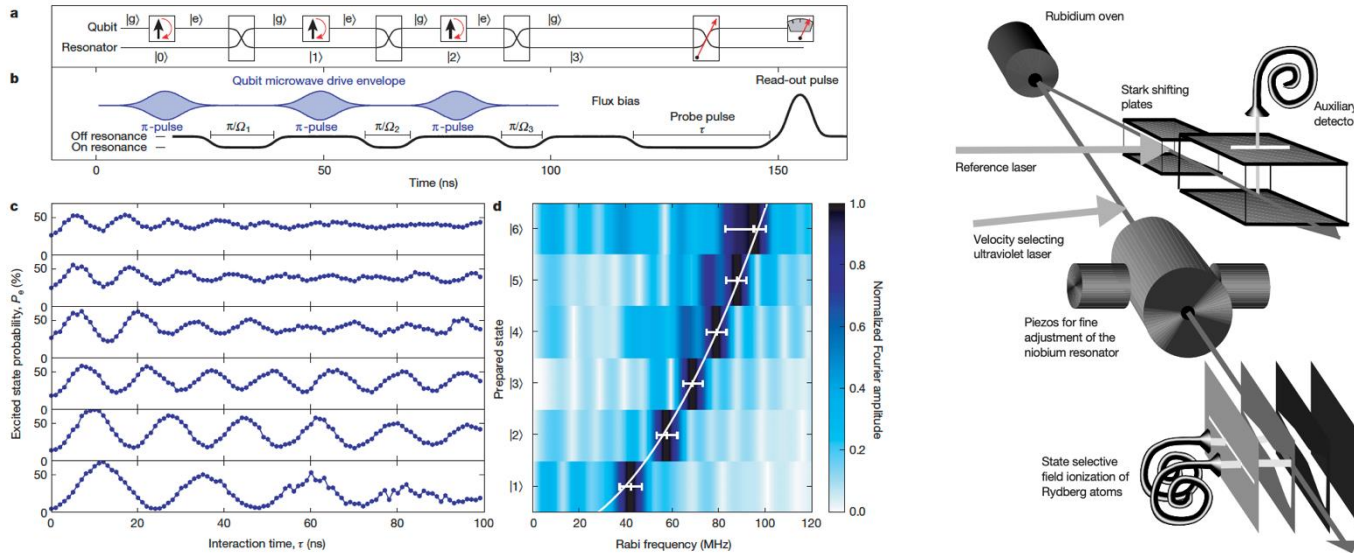
- Quantum algorithms
- Optical quantum computing
- Metrology & spectroscopy

## Fundamental

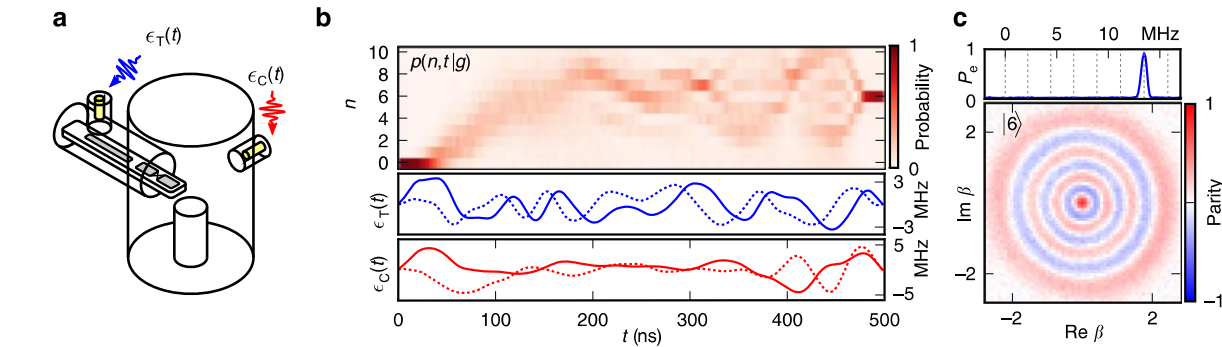
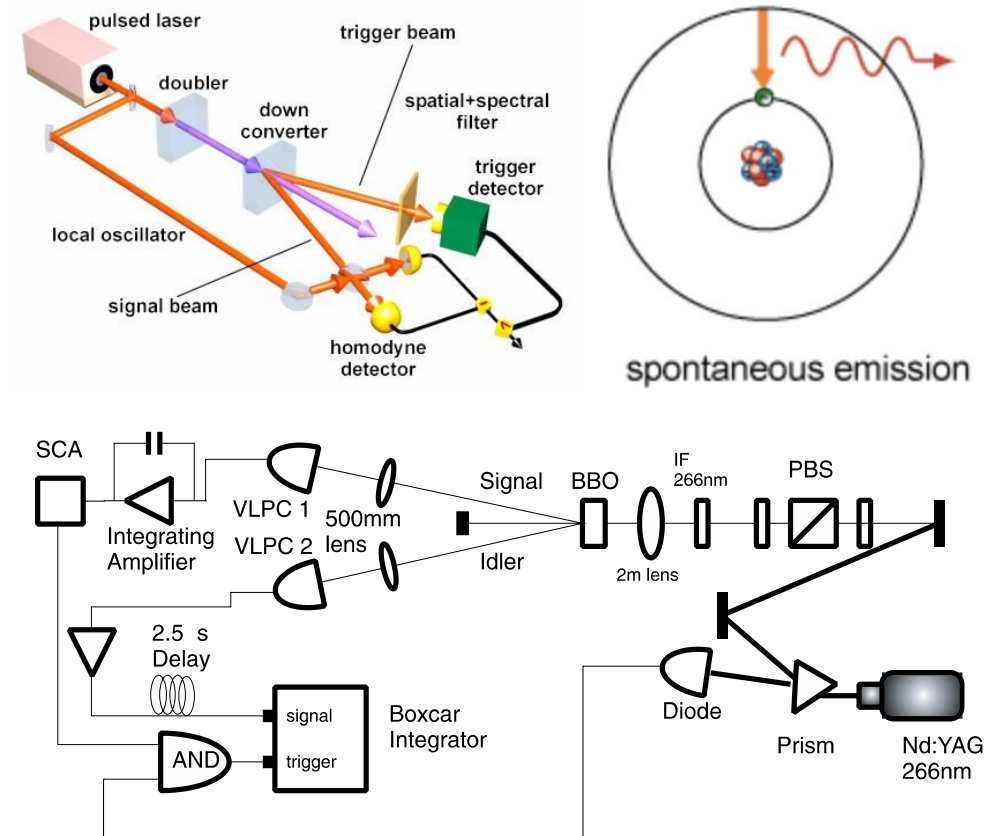
- Most basic state of light
- No classical counterpart
- Workhorse for QM states
- Macroscopic quantum light

# How Fock states are currently generated

## Microwave Fock states in resonators



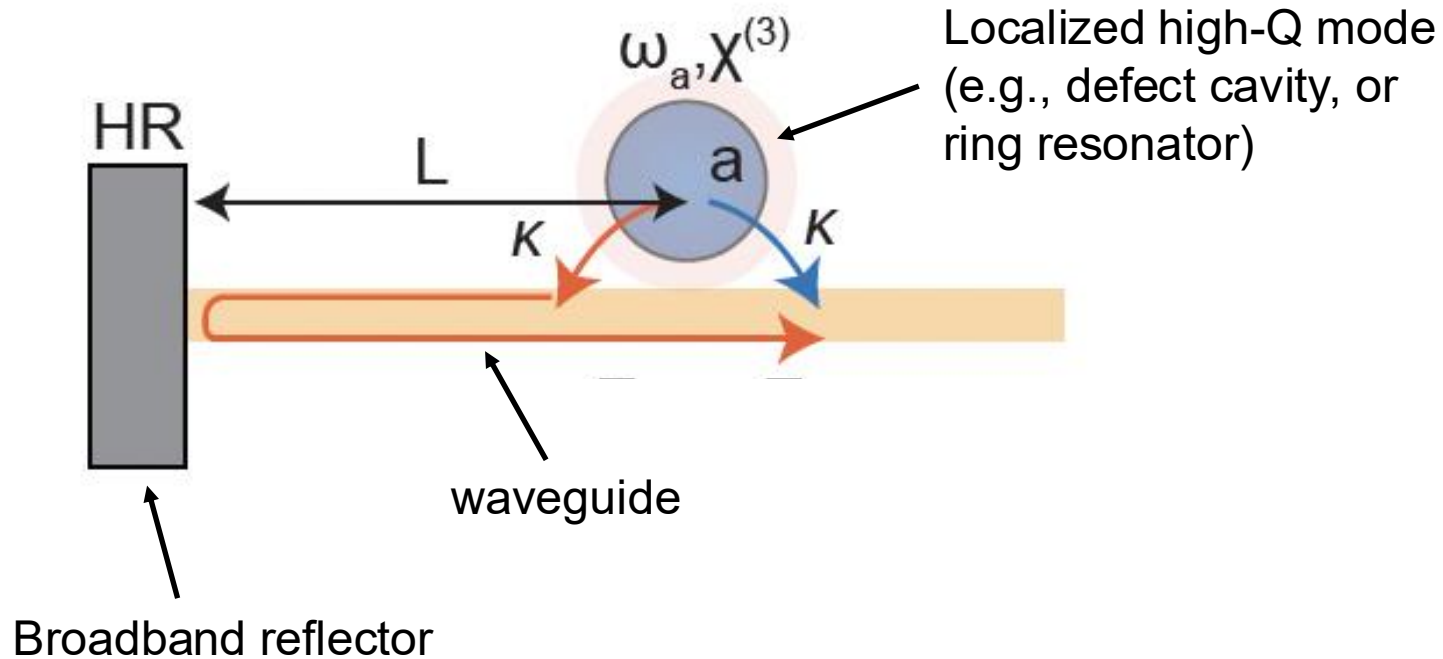
## Propagating optical Fock states



Varcoe *et al.* Nature (2000). Hofheinz *et al.* Nature (2008). Heeres *et al.* Nat. Comm. (2017). Bimbarb Nat. Phot. (2010). Waks *et al.* New J. Phys. (2006).

# Example: a nanophotonic structure with strong dependence of loss on index

## A cavity with a nonlinear resonant mirror



**Outcoupled field:**

$$\sqrt{\kappa} + \pm\sqrt{\kappa} \times e^{i\omega_a(n)L/v} \times (-i)$$

**Lossless condition:**

$$ie^{i\omega_a(n)L/v} = \pm 1$$

**Nonlinear loss:**

$$Q = Q(\omega_a(n))$$

**Frequency-dependent loss**

Hsu *et al.* Nature (2013)

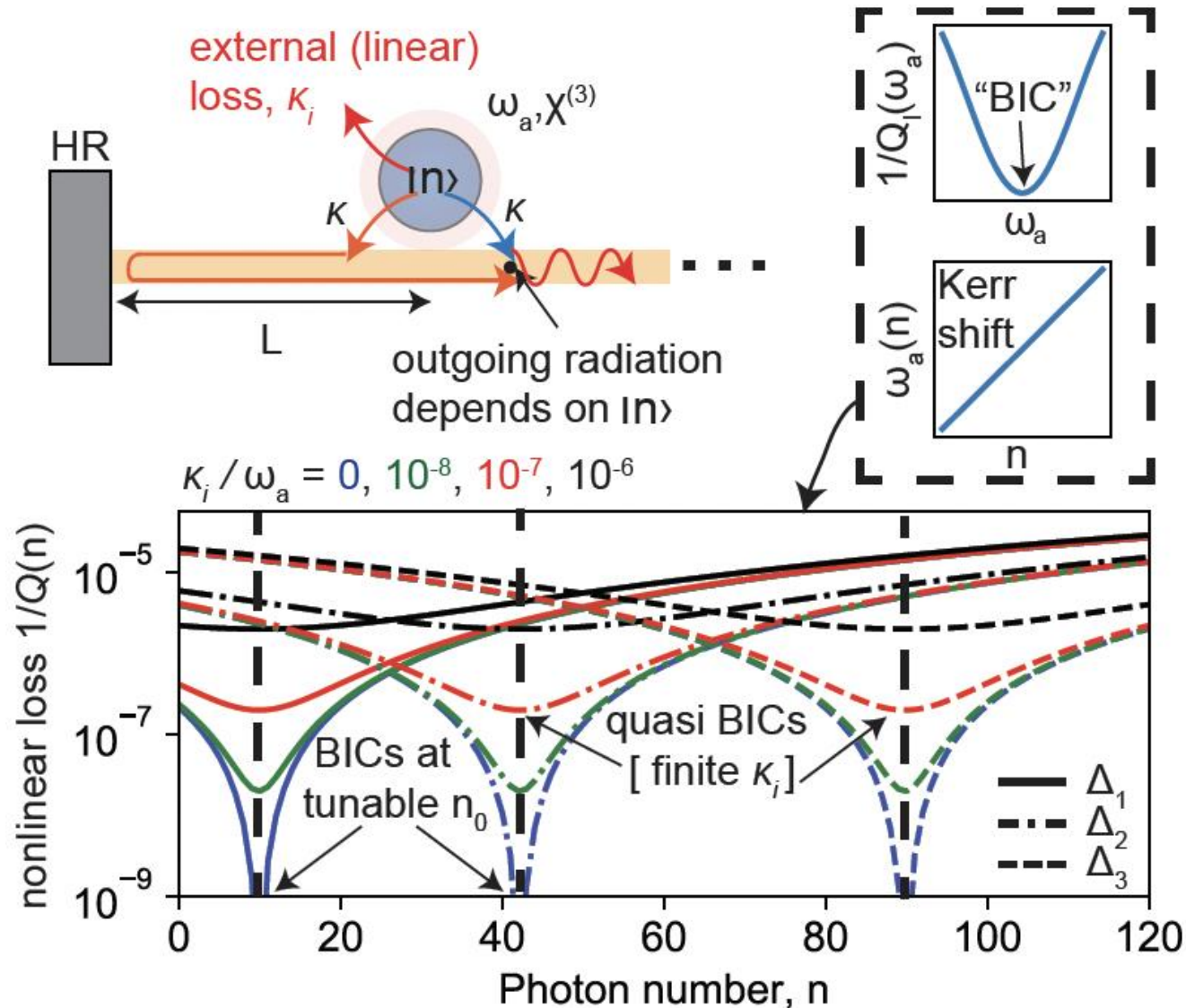
Hsu *et al.* Nat. Rev. Mat. (2016)

Yu *et al.* Nat. Phot. (2017)

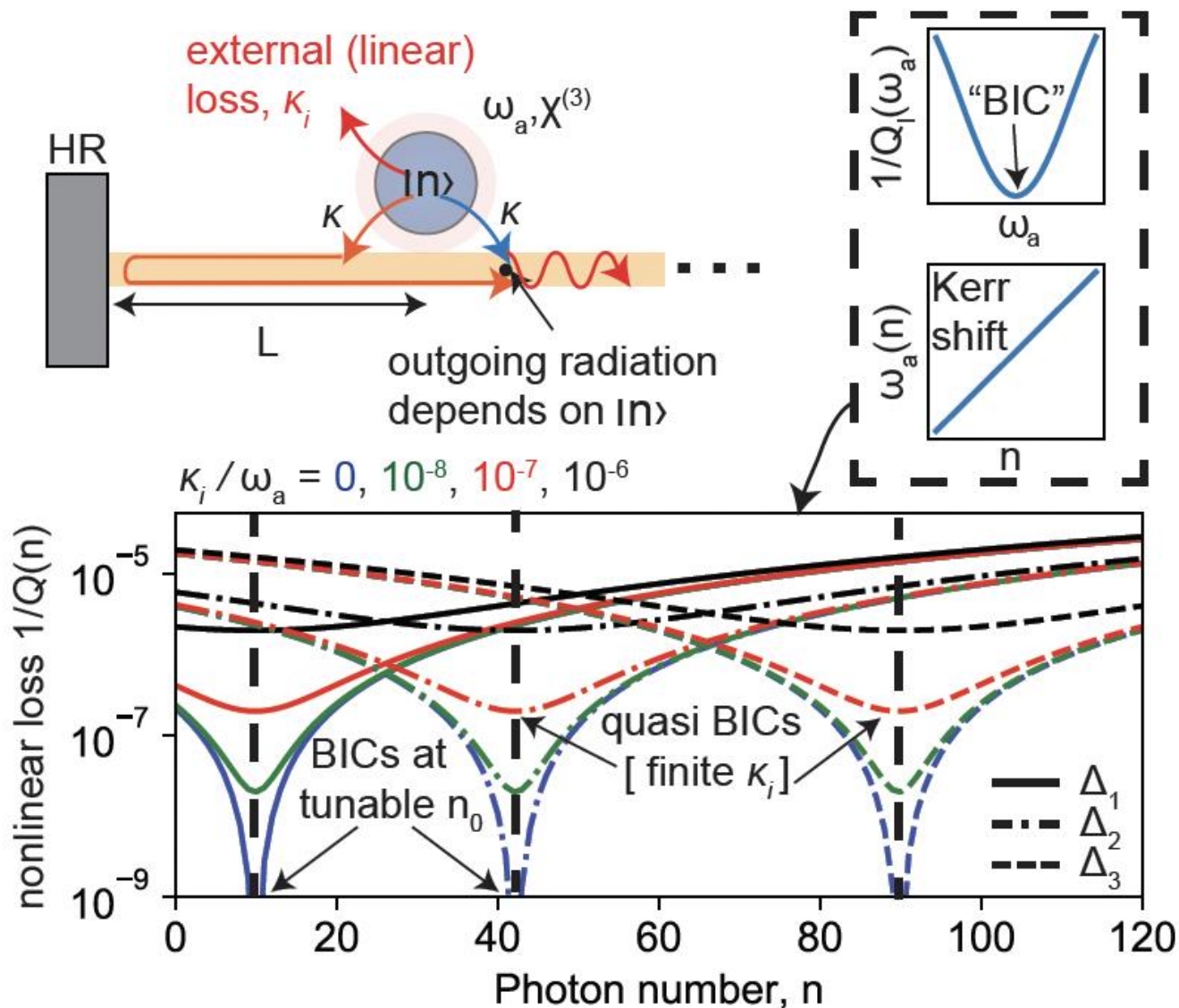
Yang *et al.* Nat. Phot. (2020)

Yu *et al.* Nat. Phot. (2021)

# Nonlinear loss and Fock stable states



# Nonlinear loss and Fock stable states



A general formalism to predict quantum state dynamics of driven-dissipative nanophotonic systems with nonlinear loss:

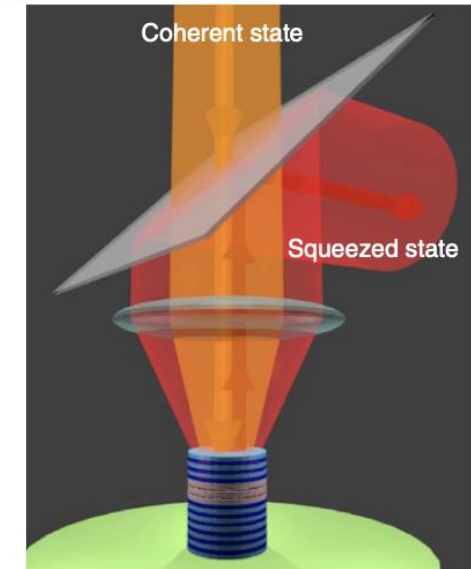
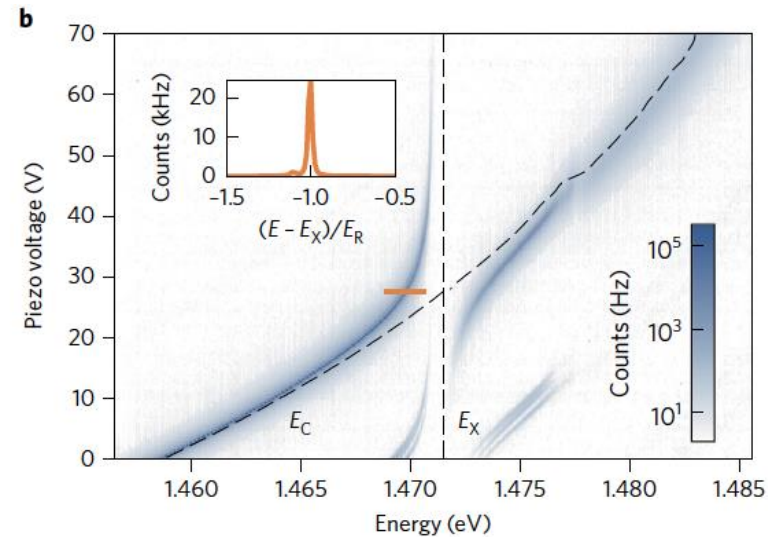
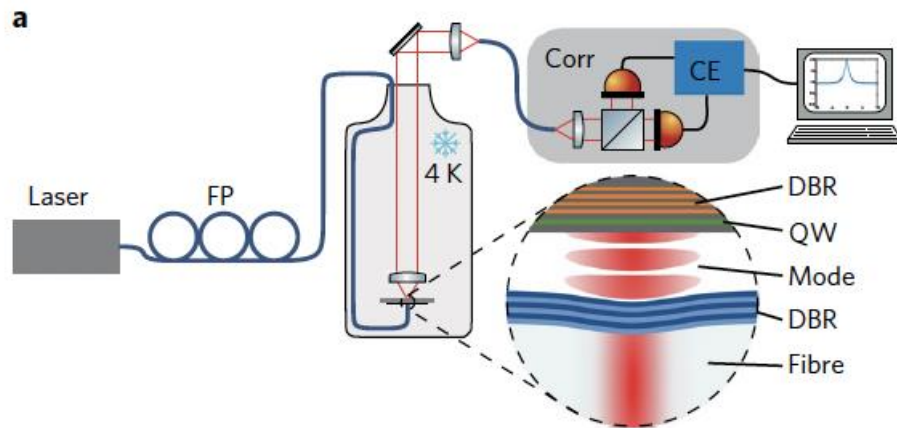
$$\dot{\rho} = -i[H_K + H_{\text{drive}}, \rho] + \mathcal{D}[\rho]$$

Nonlinear dissipator:

$$\langle m | \mathcal{D}[\rho] | n \rangle = -(mK_l(\omega_{m,m-1}) + nK_l^*(\omega_{n,n-1})) \rho_{m,n} + \sqrt{(m+1)(n+1)} (K_l(\omega_{m+1,m}) + K_l^*(\omega_{n+1,n})) \rho_{m+1,n+1}$$

Energy to add a photon to a nonlinear cavity

# Extremely strong Kerr nonlinearities from semiconductors (excitons) in microcavities



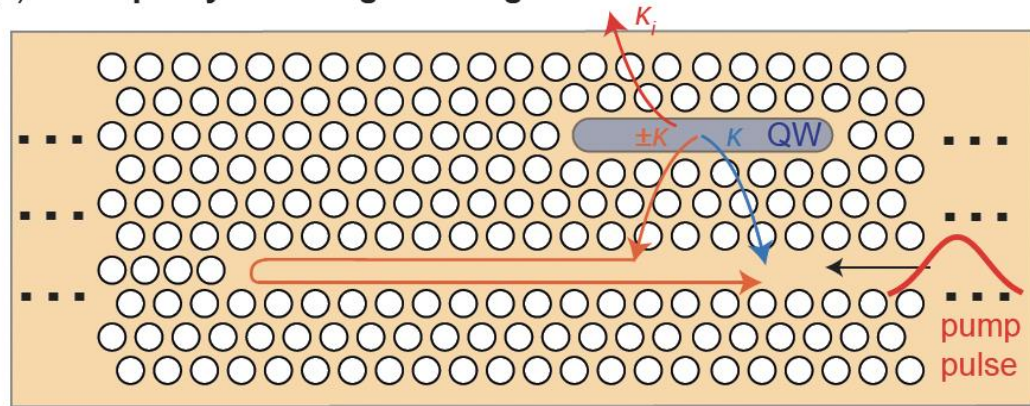
**Quantum Kerr physics in exciton-polaritons**  
Imamoglu *et al.* Physical Review Letters (1997).  
Bramati *et al.* Nature Communications (2014).  
Fink *et al.* Nature Physics (2018).  
Delteil & Fink *et al.* Nature Materials (2019).

## How close are the implementations?

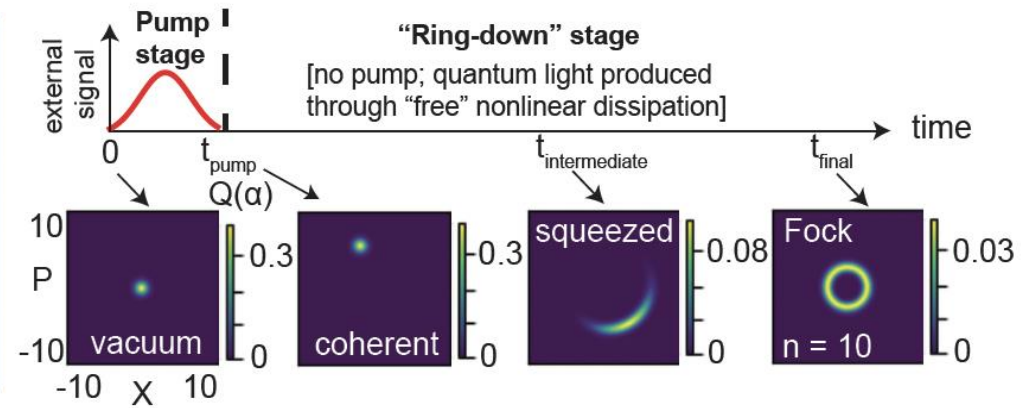
- Kerr nonlinearity within factor of 10 of loss (single-photon nonlinear regime)
- Exciton-polaritons even recently interfaced with gratings with BICs (Ardizzone *et al.* Nature (2022)).

# Prospects for observation in systems of highly nonlinear exciton polaritons

(a) Example system for generating Fock and sub-Poissonian states

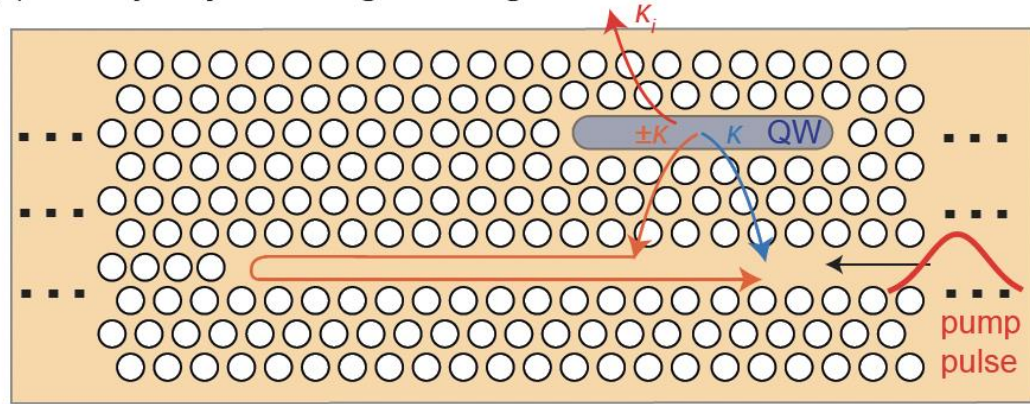


(b) Protocol for preparing Fock states

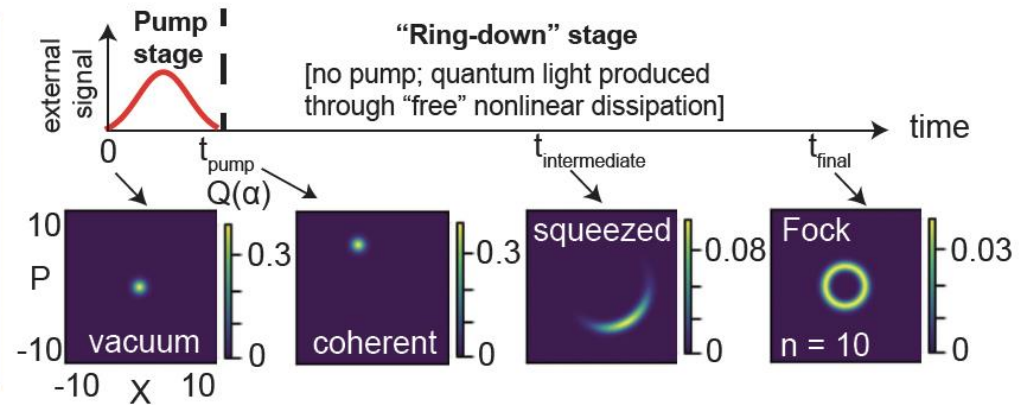


# Prospects for observation in systems of highly nonlinear exciton polaritons

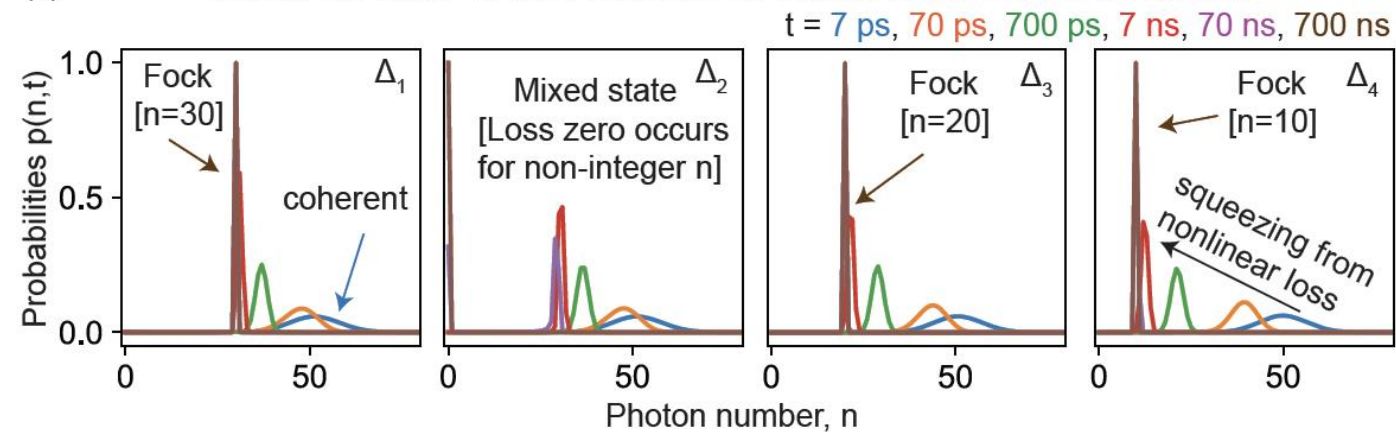
(a) Example system for generating Fock and sub-Poissonian states



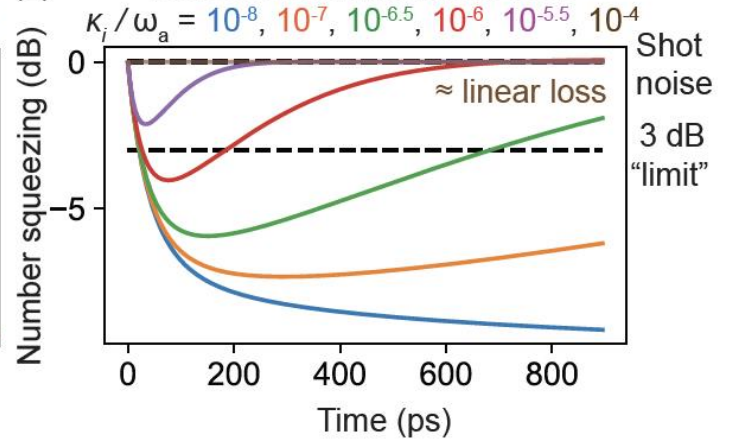
(b) Protocol for preparing Fock states



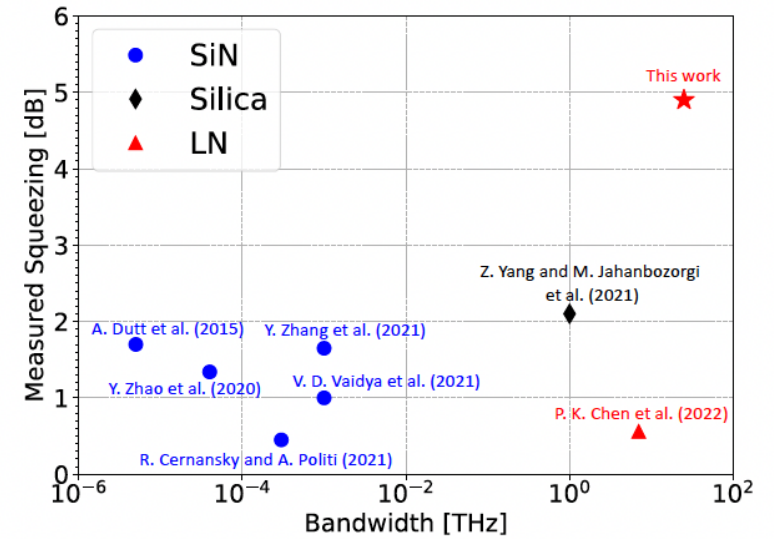
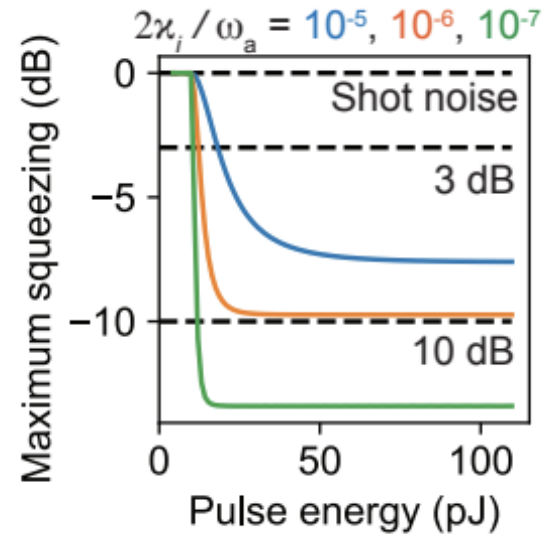
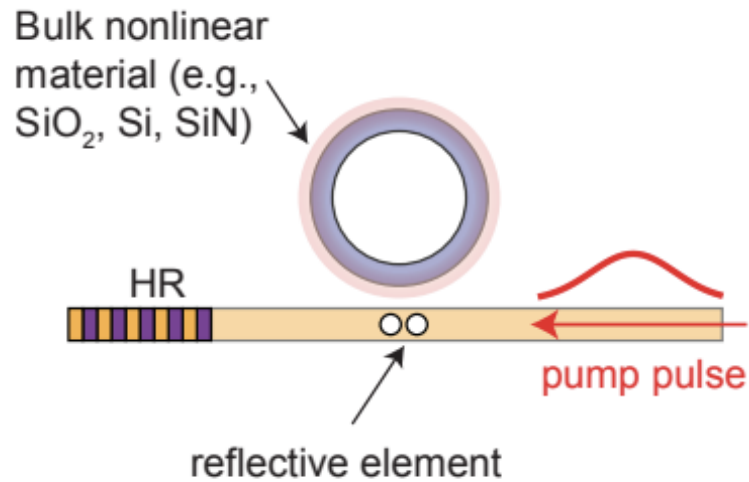
(c) Tuning the order of the Fock state by tuning the resonator frequency



(d) Effect of external linear loss



# Large squeezing in bulk nonlinear nanophotonic systems



Nehra *et al.* Science (2022).

- These effects also carry over to bulk nonlinear materials (silica, silicon, SiN, etc.)
- 10 dB squeezing (90% noise below classical shot noise level), considered a “magic number” and elusive

# A general quantum theory of outcoupling in nonlinear structures with dispersive loss

- A universal framework to predict quantum dynamics of a resonator with frequency-dependent loss.

Hamiltonian:

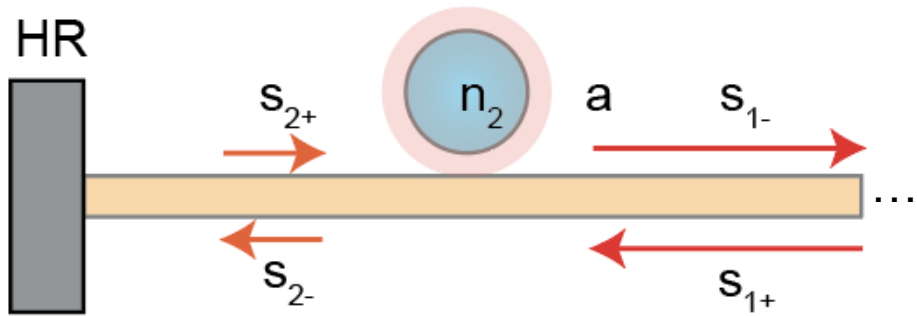
$$\begin{aligned}
 H/\hbar = & \omega_a a^\dagger a + \beta \omega_a a^{\dagger 2} a^2 + \alpha(t) a + \alpha^*(t) a^\dagger \\
 & + \sum_{i=1}^N \int \frac{d\omega}{2\pi} \omega s_i^\dagger(\omega) s_i(\omega) \\
 & + \sum_{i=1}^N \int \frac{d\omega}{2\pi} i \left( K_{c,i}(\omega) s_i(\omega) a^\dagger - K_{c,i}^*(\omega) s_i^\dagger(\omega) a \right)
 \end{aligned}$$

Dynamics of the photonic density matrix:  $\dot{\rho} = -i[H_K + H_{\text{drive}}, \rho] + \mathcal{D}[\rho]$

Nonlinear dissipator:

$$\begin{aligned}
 \langle m | \mathcal{D}[\rho] | n \rangle = & - (m K_l(\omega_{m,m-1}) + n K_l^*(\omega_{n,n-1})) \rho_{m,n} + \\
 & \sqrt{(m+1)(n+1)} (K_l(\omega_{m+1,m}) + K_l^*(\omega_{n+1,n})) \rho_{m+1,n+1}.
 \end{aligned}$$

# From frequency-dependent lifetimes to nonlinear radiation loss: an example



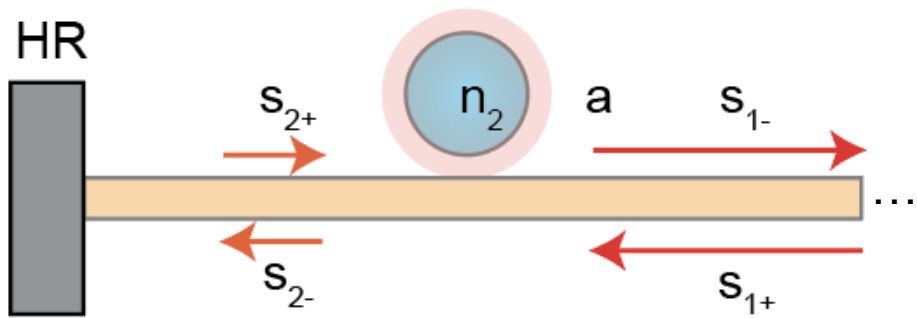
Heisenberg-TCMT with Kerr nonlinearity

$$\dot{a} = -i\omega_a a - (\kappa_1 + \kappa_2)a - i\omega_a \beta a^\dagger a^2 + \sqrt{2\kappa_1} e^{i\theta_1} s_{1+} + \sqrt{2\kappa_2} e^{i\theta_2} s_{2+}$$

Boundary conditions:

$$\begin{pmatrix} s_{1-} \\ s_{2-} \end{pmatrix} = C \begin{pmatrix} s_{1+} \\ s_{2+} \end{pmatrix} + \begin{pmatrix} \sqrt{2\kappa_1} e^{i\theta_1} \\ \sqrt{2\kappa_2} e^{i\theta_2} \end{pmatrix} a, \quad \text{and} \quad s_{2+}(\omega) = -s_{2-}(\omega) e^{i\omega T}$$

# From frequency-dependent lifetimes to nonlinear radiation loss: an example



Heisenberg-TCMT with Kerr nonlinearity

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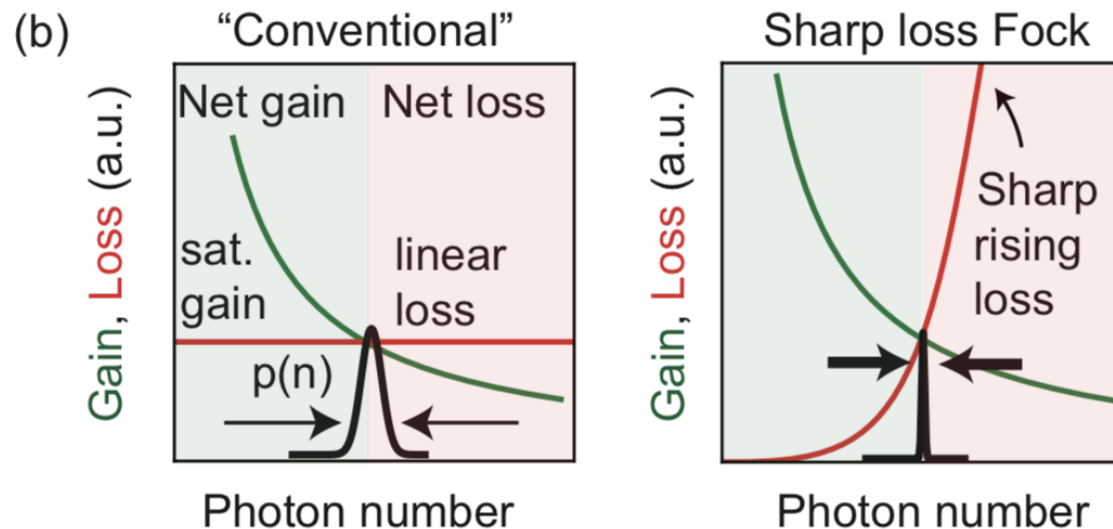
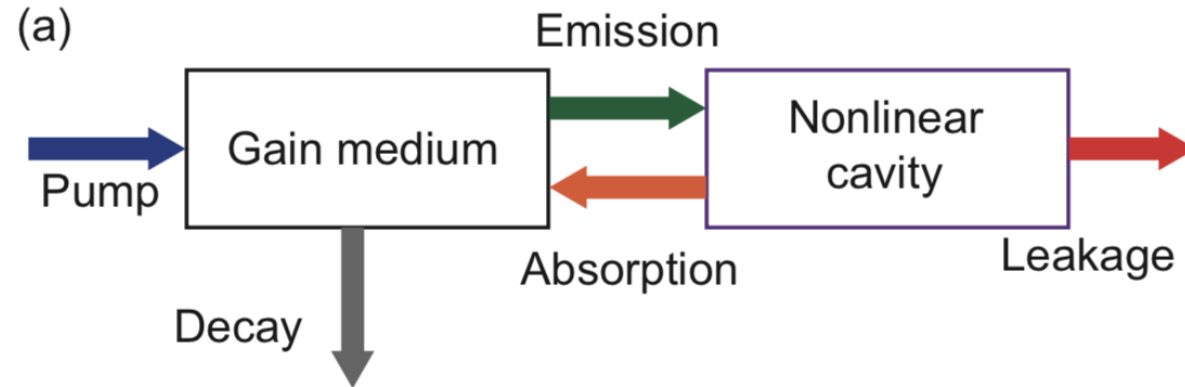
Non-Markovian equation for  $a$

$$[i(\omega_a - \omega) + K_l(\omega)] a(\omega) = K_c(\omega) s_{1+}(\omega),$$

$$K_l(\omega) = 2\kappa \left( 1 - \frac{e^{2i\theta_2}}{r_d - e^{-i\omega T}} \right) = 2\kappa \left( 1 - \frac{r_d - it_d\sigma}{r_d - e^{-i\omega T}} \right)$$

$$K_c(\omega) = \sqrt{2\kappa} e^{i\theta_1} \left[ 1 + \frac{it_d e^{i(\theta_2 - \theta_1)}}{r_d - e^{-i\omega T}} \right].$$

# Noise properties of lasers with nonlinear dissipation



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

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

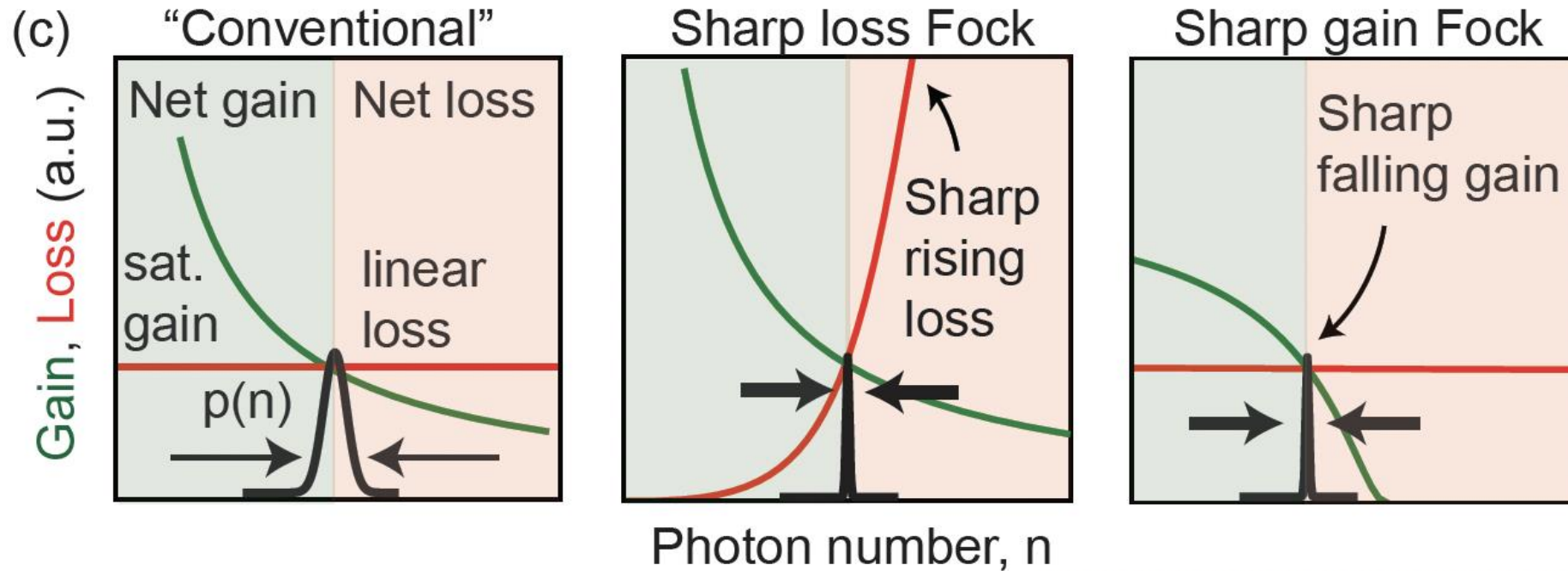
- Admits steady-state solution:

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

- Noise given by

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

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



$$p(n) \approx \frac{1}{Z} \exp \left[ -\frac{1}{2} |r'(n)| (n - \bar{n})^2 \right] \implies \Delta n = - \left( \left. \frac{d}{dn} \frac{G(n)}{\kappa(n)} \right|_{\bar{n}} \right)^{-1/2}$$

$$r(n) = G(n)/\kappa(n)$$