Terahertz photonic nonlinearity from cavity Higgs polaritons

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Abstract

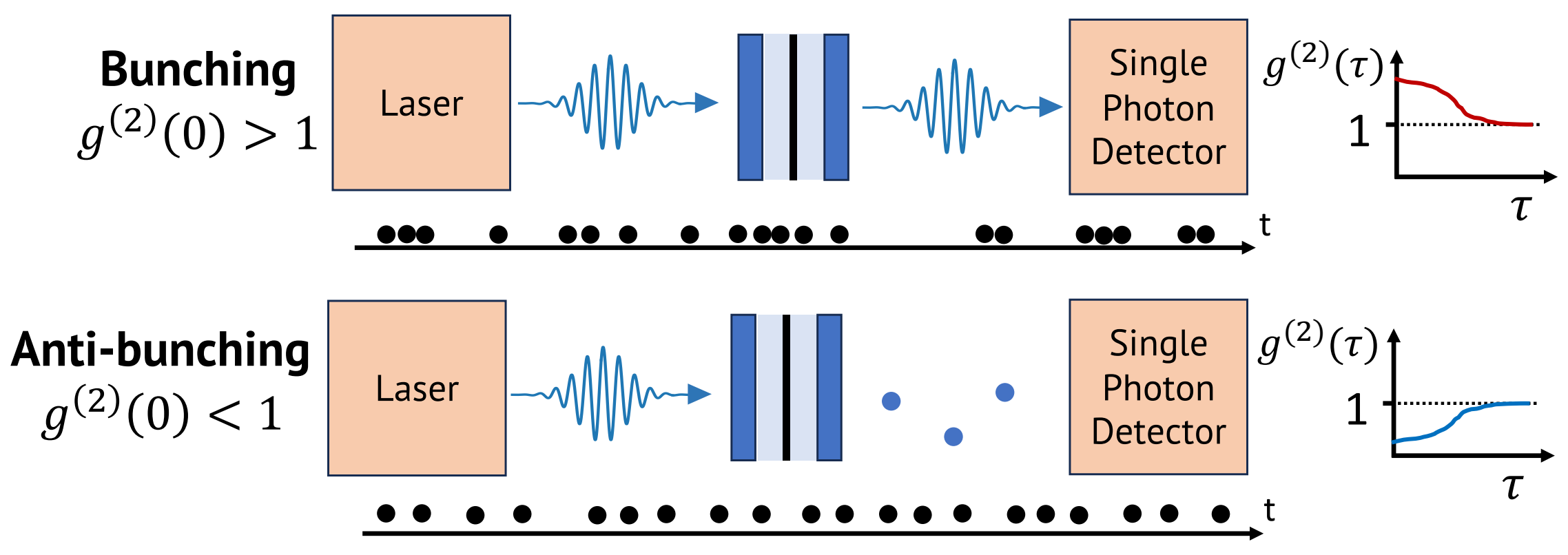
Leveraging photonic nonlinearities for the spectroscopy of low-energy matter and identification of ultrastrong light-matter coupling remain elusive. Focusing on a non-linearly coupled cavity-material system we show that as light-matter coupling increases, additional scattering pathways emerge, producing clear, testable signatures of ultrastrong coupling. Specifically, we propose a method to generate photonic nonlinearities at the single terahertz photon level by coupling a cavity to collective modes in superconductors and charge-density-wave materials. We derive a weak-coupling analytic solution and compare with the results of a non-Markovian scattering-matrix analysis at ultrastrong coupling. In doing so we reveal a diagnostic for ultrastrong coupling in the two-photon coincidence statistics that is absent in total counts.

Our Previous Works

Dissipation in quantum systems is not invariably inimical: it can be a resource to engineer desirable steady states. Dissipation can be used to engineer band structures [PRB 106, L161109 (2022)], realize non-equilibrium steady states and probe their dynamic response properties [PRB 109, 235421 (2024), npj Quantum Materials 9, 104 (2024)], and to enable a photon blockade [2411.08964].

Quantum Light

A key parameter for light is its intensity $\langle n \rangle$ and its fluctuations in intensity $\mathrm{var(n)} = \langle (a^+a)^2 \rangle - \langle a^+a \rangle^2$. It turns out that classical electromagnetism can only represent light with $\mathrm{var(n)} \geq \langle n \rangle$ corresponding to "Poissonian" or "super-Poissonian" light [1]. An equivalent experimentally measurable version of this is to consider the bunching or anti-bunching of light [2]

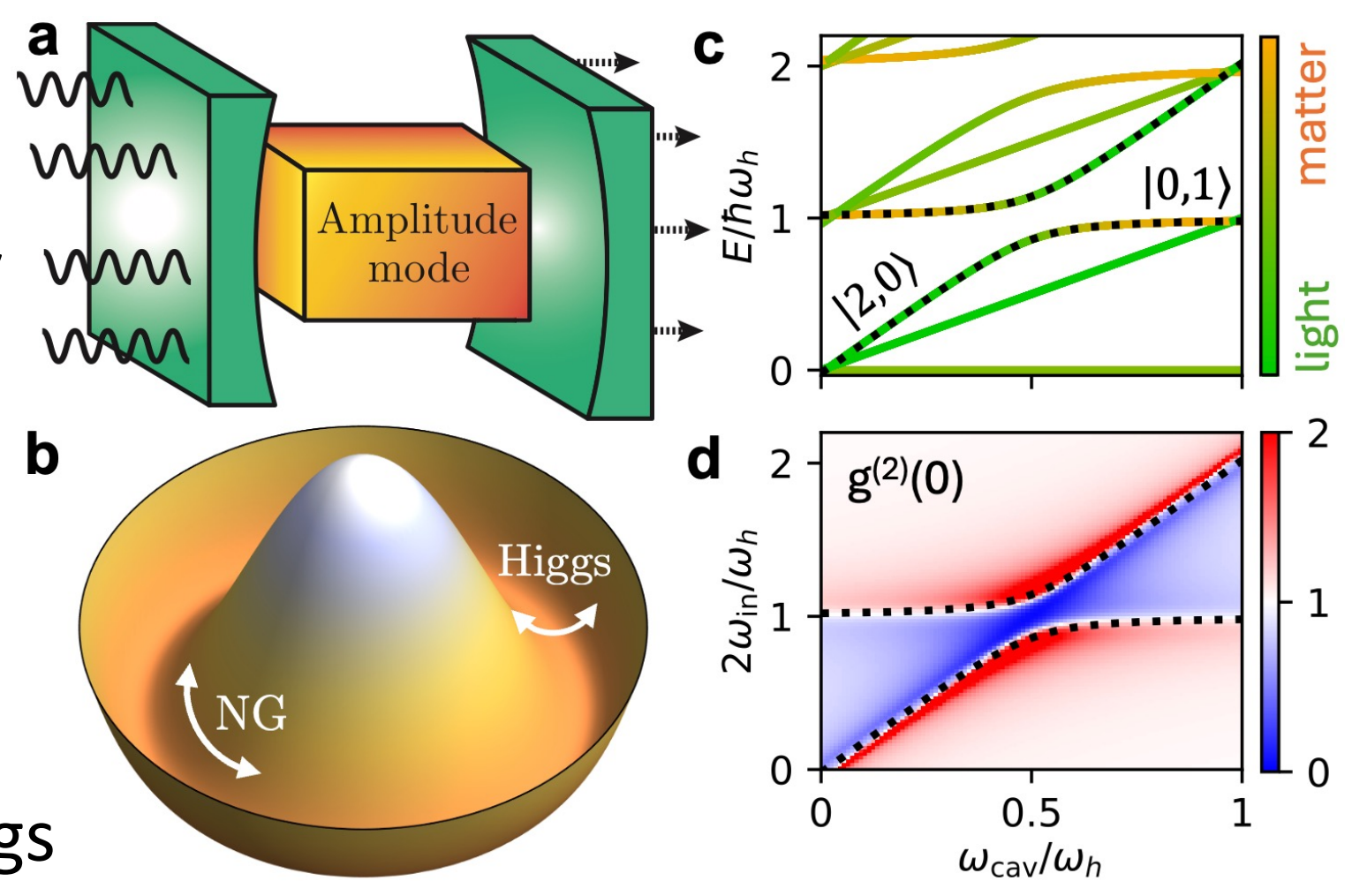


where the $g^{(2)}$ functional corresponds to intensity fluctuations. At long times intensity fluctuations are uncorrelated ($g^{(2)}$ → 1), but at short times there can be "bunching" ($g^{(2)} > 1$) and "antibunching" of light ($g^{(2)} < 1$). When $g^{(2)}(0)$ → 0 this is perfect anti-bunching and photons arrive in alone corresponding to single photon emission. The $g^{(2)}$ functional is given by

$$g^{(2)}(\tau) = \frac{\langle a^\dagger(t) a^\dagger(t+\tau) a(t+\tau) a(t) \rangle}{|\langle a^\dagger(t) a(t) \rangle|^2}$$

Higgs Polariton and Photon Blockade

System



We consider a system with a terahertz cavity coupled to an elementary collective excitation of an ordered phase, such as the Higgs amplitude mode in a superconductor or charge density wave material. We model the system with Hamiltonian

$$H_h = \hbar\omega_h h^\dagger h + \kappa(a + a^\dagger)^2 (h + h^\dagger)$$

where the cubic nonlinearity emerges from minimal substitution into a Ginzburg-Landau type model [5].

Analytic Solution

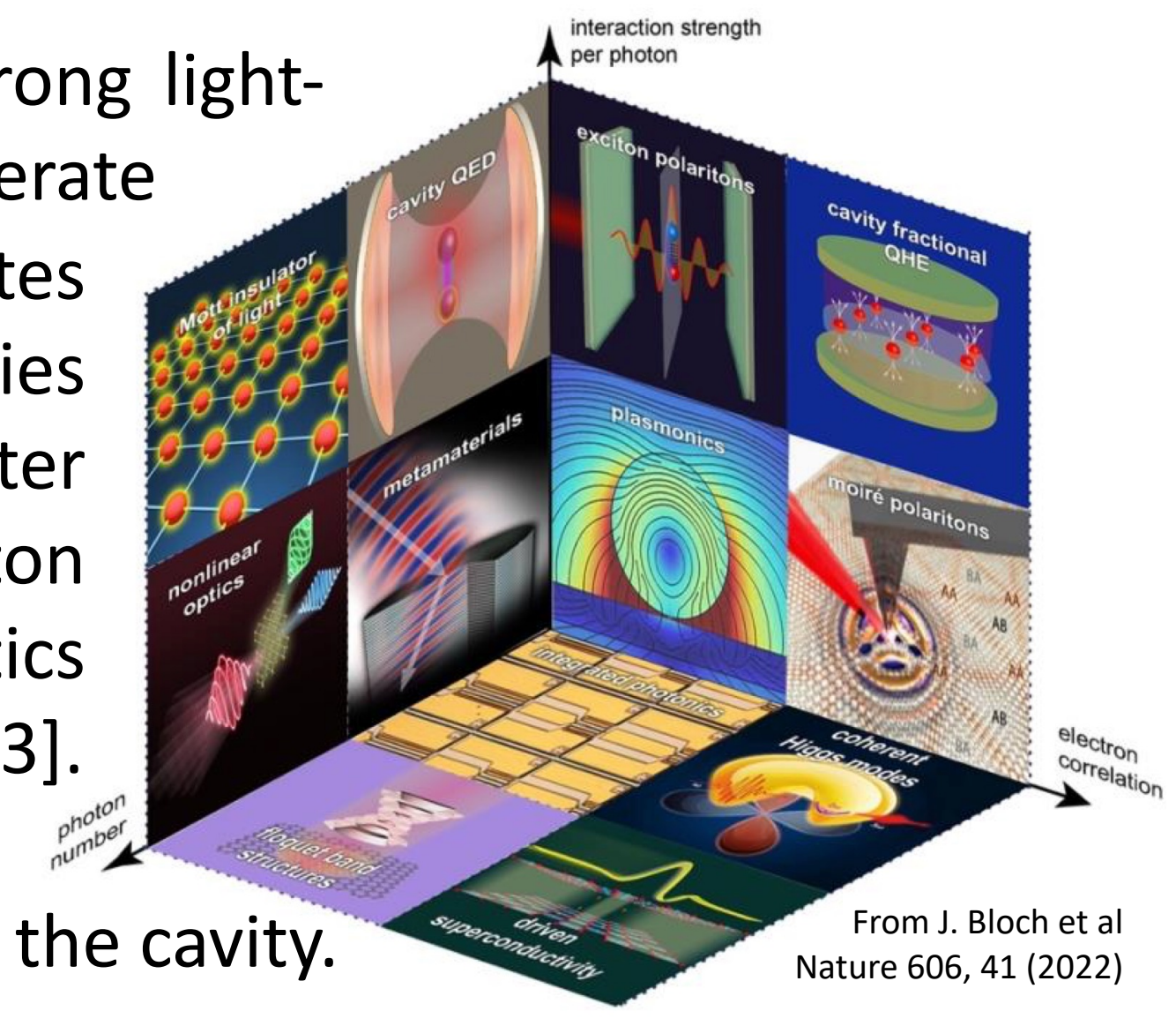
Using the P-representation we can find an analytic solution for the steady-state correlation functions of this model in the rotating wave approximation (RWA) where we approximate the nonlinearity with $\kappa(aah^+ + a^+a^+h)$ and dissipation is introduced via Lindblad dissipators $J_a = \sqrt{\gamma_{\mathrm{cav}}}a$ and $J_h = \sqrt{\gamma_h}h$ [6]

$$g^{(2)}(0) = \left| \frac{(\gamma_{\mathrm{cav}} + i(\omega_{\mathrm{cav}} - \omega_{\mathrm{in}}))(\gamma_h + i(\omega_h - 2\omega_{\mathrm{in}}))}{\kappa^2 + (\gamma_{\mathrm{cav}} + i(\omega_{\mathrm{cav}} - \omega_{\mathrm{in}}))(\gamma_h + i(\omega_{\mathrm{cav}} - 2\omega_{\mathrm{in}}))} \right|^2$$

Polariton Induced Photon Blockade

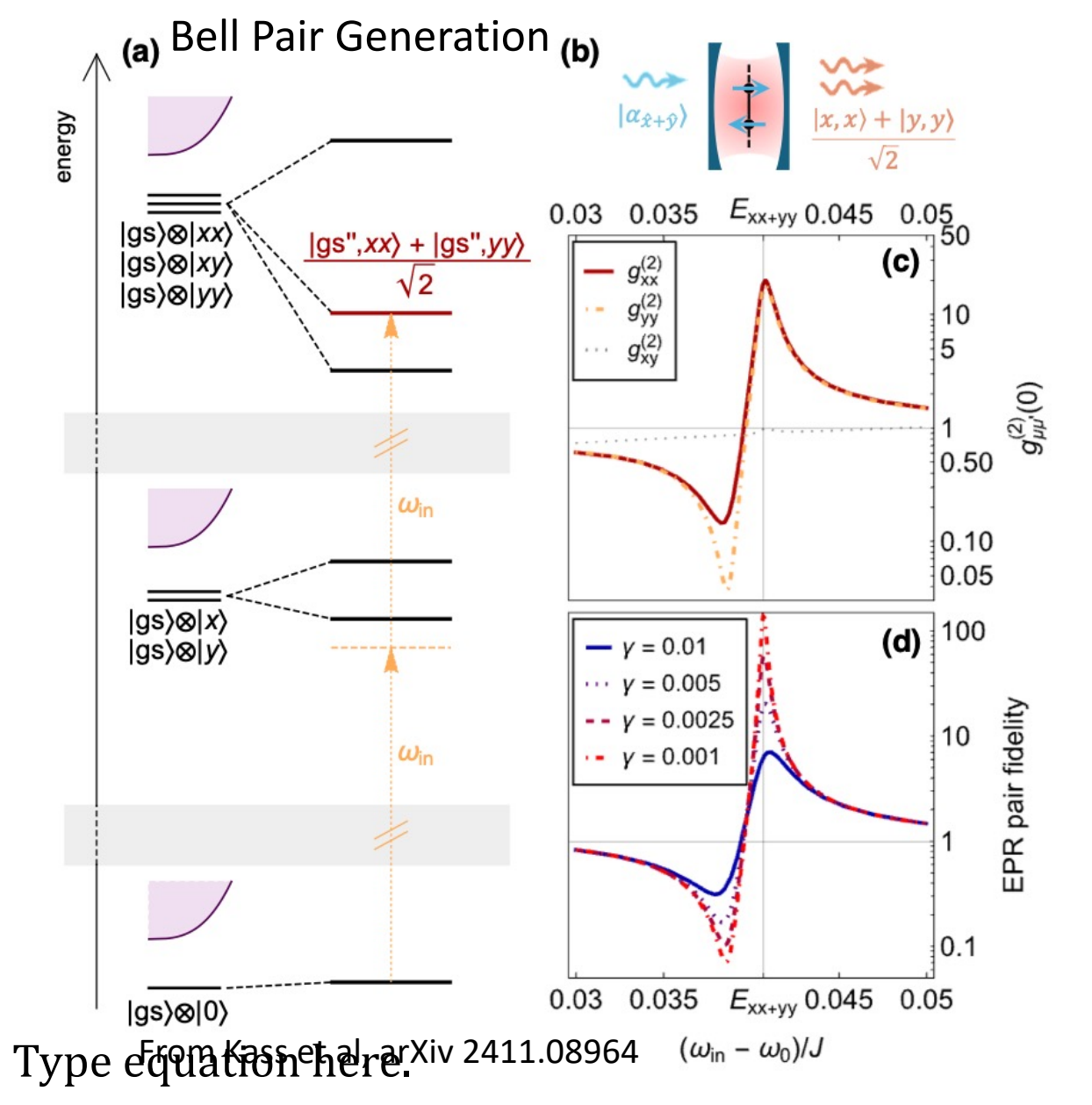
Coupling matter modes to light creates polaritonic mixtures of light and matter near avoided crossings. For superconducting modes coupled to photons in a cavity this is a Higgs polariton [7]. The splitting at the anti-crossing enables single photon generation: the one-photon resonance is not half the energy of the two-photon resonance and so there is a photon blockade when $2\omega_{\mathrm{cav}}$ is in the gap. We see this in panels (c) and (d) above where antibunching is in the polaritonic gap.

Cavity Quantum Materials



From J. Bloch et al
Nature 606, 41 (2022)

Cavities can enable strong light-matter coupling to generate hybrid light-matter states with distinct properties from the light or matter alone such as polariton physics, non-linear optics and superconductivity [3]. Input/output light is used to drive/dissipate the cavity.



From Kass et al. arXiv 2411.08964

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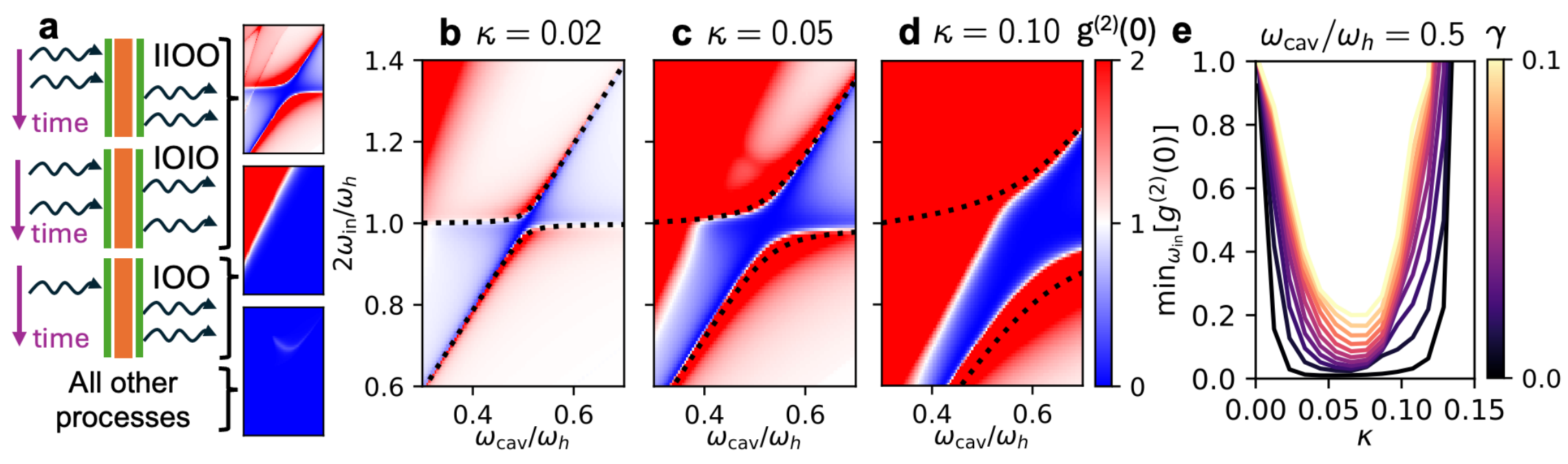
Here we take a different approach by focusing on controlling the output state of light using a cavity-material system. In our previous work [4] we showed that optical Raman processes of a material near a critical point can emit single photons and Bell pairs.

New Features at Ultrastrong Coupling



At weak light-matter coupling strength the dominant contribution to (anti)bunching is resonant absorption and reemission of two photons. Once light-matter coupling becomes ultrastrong the counter-rotating terms become significant, the cavity acquires a dark occupation and a dispersive coupling term, $2a^+a(h+h^+)$ is relevant. As a result, new features including stimulated emission arise which can be used to diagnose ultrastrong coupling even when the transmission is similar to that of weak coupling.

Scattering Matrix Theory

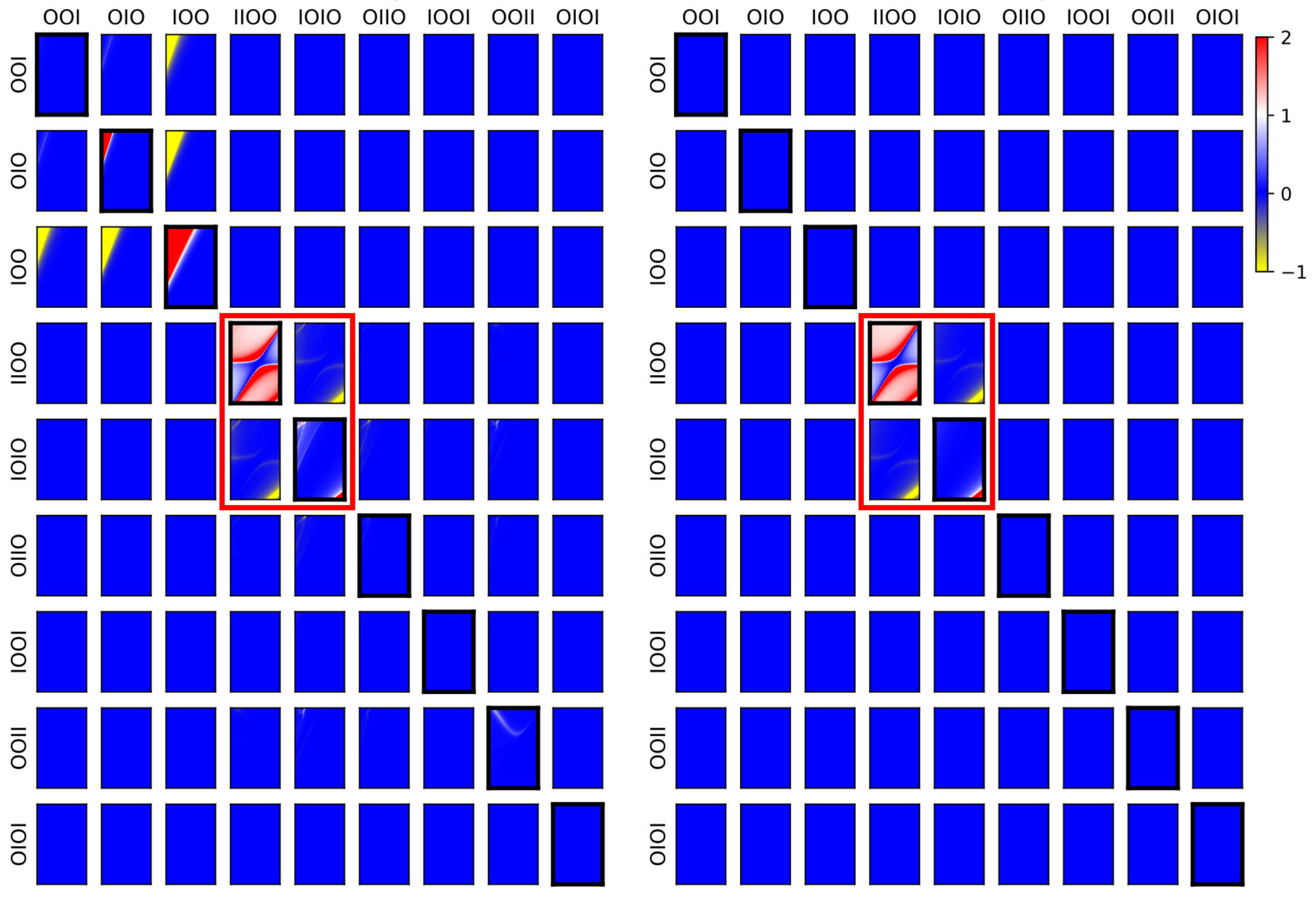
To compute photon statistics at ultrastrong coupling we use a non-Markovian scattering matrix approach [8] which breaks into pathways N_i (up to frequency integrals)

$$g^{(2)} = \sum\nolimits_i |\sum\nolimits_f \langle f | N_i | 0 \rangle|^2$$

For example, the in-in-out-out process is (with $\mathcal{G}(E) = (E - H - \Sigma^R)^{-1}$ & self-energy $\Sigma^R = -i\gamma_a a^+a - i\gamma_h h^+h$)

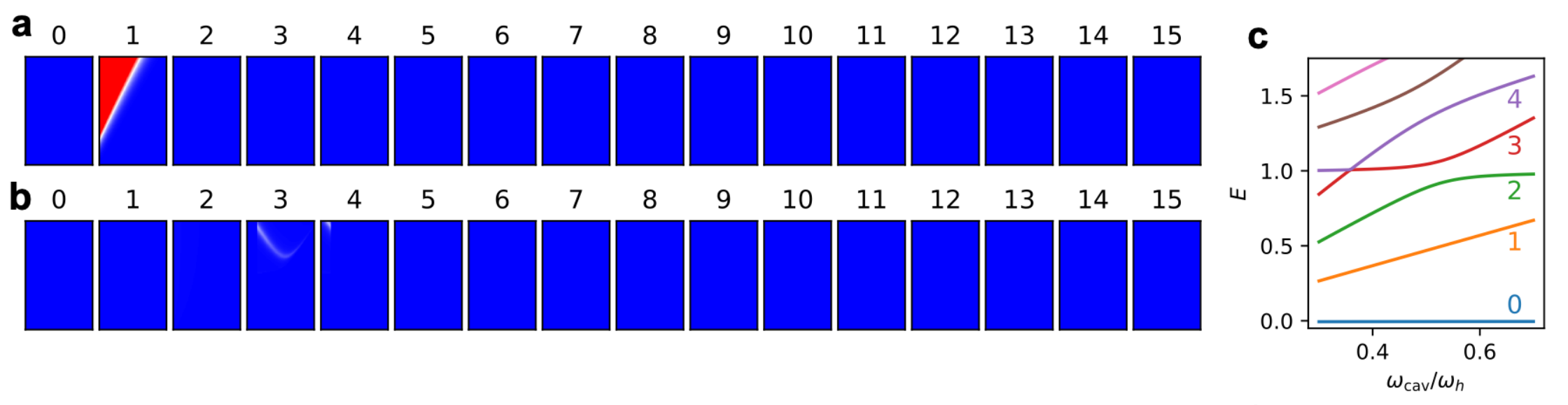
$$N_{IIOO} = a\mathcal{G}(E_0 + 2\omega_{\mathrm{in}} - \hbar\omega) a \mathcal{G}(E_0 + 2\omega_{\mathrm{in}}) a^\dagger \mathcal{G}(E_0 + \omega_{\mathrm{in}}) a^\dagger$$

Decomposing panel (c) we see terms appear beyond RWA.



Stimulated Emission and Parity

We can further decompose the features not just by their scattering pathways, but we can account for their final states too. Doing so we see that two of the new features at ultrastrong coupling end in states other than the ground state. (a) Stimulated emission ends in the 1 state which is mostly $|1{,}0\rangle$ in the basis $|n_{\mathrm{cav}}, n_{\mathrm{h}}\rangle$. (b) The weak feature ends in the upper polariton state.



Considering the bosonic parity operator $e^{i\pi\, a^+a}$ which separates states into even (+) and odd (−) sectors we see that the IOO process we must go +−+−, while for the OOII process we must go +−+−+ since the Hamiltonian preserves parity. The ground state is an even parity eigenstate and so it can be written as

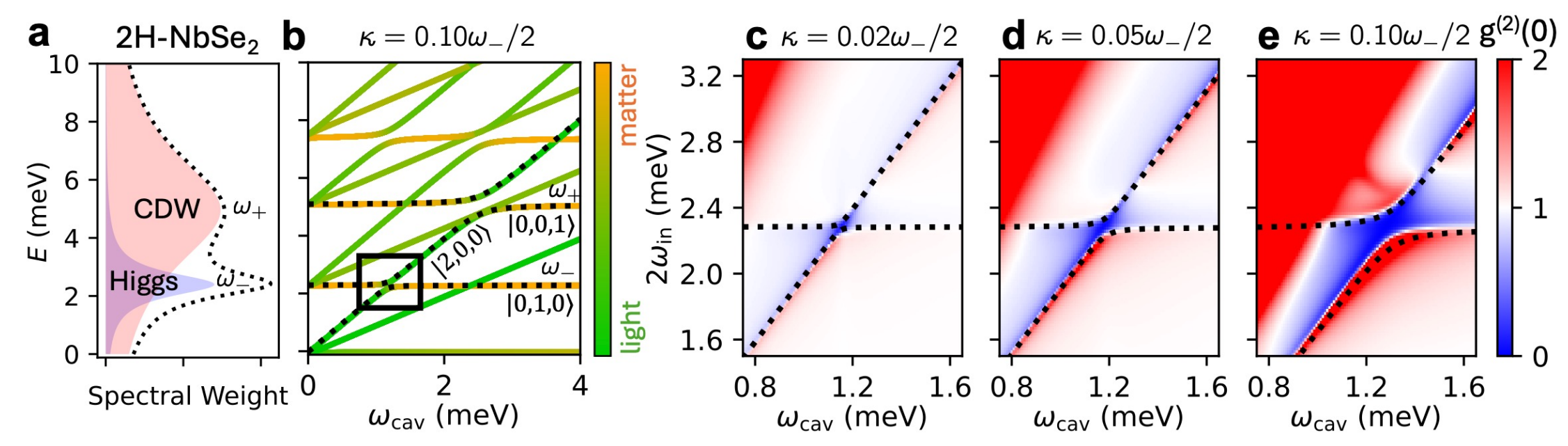
$$|0\rangle = \sum\nolimits_{n_{\mathrm{cav}}\ \mathrm{even}, n_h} c_{n_{\mathrm{cav}}, n_h} |n_{\mathrm{cav}}, n_h\rangle$$

Where the scattering pathways for (a) and (b) include

$$|2,0\rangle \overset{a^\dagger}{\to} |3,0\rangle \overset{a}{\to} |2,0\rangle \overset{a}{\to} |1,0\rangle$$

$$|2,0\rangle \overset{a}{\to} |1,0\rangle \overset{a}{\to} |0,0\rangle \overset{a^\dagger}{\to} |1,0\rangle \overset{a^\dagger}{\to} |f\rangle = |2,0\rangle \sim |UP\rangle$$

Applications to 2H-NbSe$_2$



Now, the analysis presented above applies to amplitude modes in general. Making this more specific we consider 2H-NbSe2 where superconductivity and CDW coexist. This makes the Higgs more visible and we model it with

$$H = \hbar\omega_{\mathrm{cav}} a^\dagger a + \hbar\omega_h h^\dagger h + \hbar\omega_c c^\dagger c + \kappa_{hc}(h^\dagger c + c^\dagger h)$$
$$+ \kappa_h (a + a^\dagger)^2 (h + h^\dagger) + \kappa_c (a + a^\dagger)^2 (c + c^\dagger)$$

Plotting, we see that many of the features from the single mode model persist in the model with two modes. In addition, we find that the antibunching due to the CDW overlaps with the antibunching from the Higgs mode. Other candidate systems exist too.

Type	Material	T_c	$\hbar\omega_{\mathrm{mode}}/2$
SC	2H-NbSe$_2$	7.2 K	1.2 meV
SC	2H-TaS$_2$^{a}	6.4 K	0.62 meV
Bulk CDW	1T-TaS$_2$	200 K	5.0 meV 7.1 meV
Bulk CDW	1T-TiSe$_2$	202 K	4.8 meV 7.4 meV
1D CDW	o-TaS$_3$	220 K	1.1 meV
1D CDW	(TaSe$_4$)$_2$I	263 K	5.6 meV
1D CDW	(NbSe$_4$)$_{10}$I$_3$	289 K	6.3 meV

^{a} At 6 GPa

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