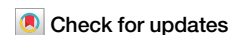


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Raman-phonon-polariton condensation in a transversely pumped cavity

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Phonon polaritons are hybrid states of light and matter that are typically realised when optically active phonons couple strongly to photons. We suggest a new approach to realising phonon polaritons, by employing a transverse-pumping Raman scheme, as used in experiments on cold atoms in optical cavities. This approach allows hybridisation between an optical cavity mode and any Raman-active phonon mode. Moreover, this approach enables one to tune the effective phonon–photon coupling by changing the strength of the transverse pumping light. We show that such a system may realise a phonon-polariton condensate. To do this, we find the stationary states and use Floquet theory to determine their stability. We thus identify distinct superradiant and lasing states in which the polariton modes are macroscopically populated. We map out the phase diagram of these states as a function of pump frequencies and strengths. Using parameters for transition metal dichalcogenides, we show that realisation of these phases may be practicably obtainable. The ability to manipulate phonon mode frequencies and attain steady-state populations of selected phonon modes provides a new tool for engineering correlated states of electrons.

Phonon polaritons result from strong coupling between light and lattice vibrations, leading to new normal modes with hybrid properties of both constituents¹. Because phonons play an essential role in material properties—including mediating phases such as superconductivity—manipulating phonon properties by coupling to light is an intriguing prospect². However, phonons are typically in the terahertz (THz) or infrared range, and direct strong coupling requires cavities supporting THz or infrared cavity modes. Moreover, only optically active phonon modes—i.e., those with a dipole matrix element—can couple directly to light. In many cases, relevant phonon modes may not be optically active; see e.g.³. As we demonstrate further below, this restriction can be overcome by using a Raman pumping scheme. That is, one couples to the phonons via a two-photon transition involving cross terms between cavity photons and an external pump. This engineers an effective coupling between optical cavity photons and terahertz phonons.

Raman-driving schemes⁴ have proven highly fruitful for experiments involving cold atoms in optical cavities. Crucially, such schemes realise a scenario that may be referred to as “synthetic cavity QED,” in which one can control the strength of matter–light coupling as well as the effective energies of the matter and light degrees of freedom. Following the original proposal⁴, this enabled the realisation⁵ of the Dicke–Hepp–Lieb superradiance transition^{6–8}. Further extensions of this core idea of Raman driving to multimode confocal cavities⁹ have introduced tunable-range¹⁰ and sign-changing interactions¹¹ to cavity QED, enabling the realisation of an optical

lattice with sound modes¹² and replica-symmetry breaking in a spin glass¹³. See the reviews^{14,15} for further discussion. Our aim in this manuscript is to explore how the power and flexibility of Raman-driving can be extended to solid-state systems.

As noted above, being able to manipulate the properties of solid-state phonon modes is of significant interest due to the possibility of engineering correlated electronic phases. Using transient light pulses to modify phonon modes has been central to proposals for, and realisations of, light-induced phase transitions in cuprates and other superconductors^{3,16–18}. Such results rely on direct, terahertz coupling of light to the phonons in the transient, pulsed-laser regime, and this inevitably causes heating while posing challenges to those attempting to interpret the results. This has led to proposals to instead use strong coupling to infrared or THz cavities^{19,20}, including a variety of proposals of how cavity QED may enhance superconductivity^{21–23}. Even with a cavity, the coupling is restricted to optically active modes. To overcome this, Raman driving schemes are required; see Ref. 24 for an alternative Raman scheme providing parametric coupling between a THz photon and phonon.

In this paper, we show that it is possible to achieve a superradiant phase—i.e., a phonon-polariton condensate—using a continuous-wave Raman driving scheme coupling a phonon to an optical-frequency cavity mode. We present a model for independent, driven excitons in an optical cavity and use the mean-field ansatz to show that sufficiently strong driving leads to an

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instability of the normal phase, understood to be the phase with no macroscopic population of the cavity. In contrast to Raman-driving schemes for cold atoms, we find that adiabatic elimination of the exciton states is not quantitatively accurate for the parameters relevant to the materials we study in this work. This requires us to directly model the periodically driven system, and to use Floquet analysis to understand the stability of states.

We show that the instability can take two forms, either due to super-radiance or due to lasing, and we describe the features of each. We present the phase diagram for the model and discuss the effect of different parameters. Our model is inspired by transition metal dichalcogenides (TMDs), particularly TMD monolayers; these have attracted much attention for their optoelectronic properties²⁵. Key to the results below is that they host sharp²⁶, strongly bound, optically active excitons (electron-hole pairs)^{27,28} that couple to sufficiently long-lived optical phonons with a Huang-Rhys parameter of approximately unity^{29,30}. The model presented in this paper is, however, not limited to TMD materials, but presents a general approach for how Raman-driving may be used in solid-state contexts. Hence the results below may be applied to more exotic few-layer and twisted multilayer materials.

Results

Model: phonons and driven excitons in a cavity

We consider a model that involves the interaction of cavity photons $\hat{a}$, excitons $\hat{X}_j$, and phonons $\hat{b}_j$. Our model considers explicit exciton-photon coupling (including optical driving of the excitons by the pumps) and exciton-phonon coupling. As discussed further below, this leads to an effective Raman coupling between phonons and photons; however, in contrast to other Raman-driven schemes⁴, we will not adiabatically eliminate the excitons.

To simplify the calculation, we consider the case of a single-mode optical cavity. We will remark later on how these results might change in a multimode cavity. Furthermore, we consider a model of quasilocalsed excitons on sites labelled j , each with an associated phonon mode. Such a model is an approximation to the full description of TMD materials, in which both excitons and phonons are delocalised. However, such a model is a reasonable first approximation when considering excitons and phonons with relatively flat dispersions, which can be the case for TMDs^{28,31}. Setting $\hbar = 1$ throughout, this model takes the form $\hat{H} = \omega_c \hat{a}^\dagger \hat{a} + \sum_{j=1}^N \hat{H}_{\text{mat},j}$, where the matter Hamiltonian for each site j is

$$\hat{H}_{\text{mat},j} = \omega_{\text{ex}} \hat{X}_j^\dagger \hat{X}_j + \omega_{\text{phon}} \left[\hat{b}_j^\dagger \hat{b}_j + \Delta q (\hat{b}_j^\dagger + \hat{b}_j) \hat{X}_j^\dagger \hat{X}_j \right] + (\hat{X}_j^\dagger + \hat{X}_j) \left[\Omega_+ \frac{e^{i\omega_+ t}}{2} + \Omega_- \frac{e^{i\omega_- t}}{2} + g_0 \hat{a}^\dagger + \text{H.c.} \right]. \quad (1)$$

We note that Δq parameterises the exciton-phonon coupling, which takes the Holstein form³². This Holstein form may be developed from electron- and hole-phonon couplings and has been used to model TMD exciton lineshapes^{33,34}. The Huang-Rhys parameter S ³⁵, which is typically used to define coupling to phonons, is related to Δq by $S = \Delta q^2$. Such coupling to vibrational modes is what leads to Raman scattering, so our discussion is focused on Raman-active phonon modes. The terms $\Omega_\pm e^{i\omega_\pm t}$ denote the two driving fields. As discussed elsewhere^{4,36}, two-frequency driving is used to ensure resonance of both co- and counter-rotating couplings between photons and phonons, as illustrated in Fig. 1. That is, there are two processes we want to make resonant: The process that destroys a photon and creates a phonon (the co-rotating process), and the process that creates both a photon and a phonon (the counter-rotating process). When using a single pump frequency, the pump can be chosen to make one of these two-photon transitions resonant, but not both. Using two pumps allows for the simultaneous two-photon resonance of both processes. As we will discuss further below, this simultaneous resonance can reduce the threshold power required by absorbing the phonon energy into the pumps' difference frequency.

In addition to the Hamiltonian, we consider effects of dissipation by considering a Lindblad master equation³⁷: $i\partial_t \rho = [\hat{H}, \rho] + i\sum_j \mathcal{D}[\hat{L}_j]$, with $\mathcal{D}[\hat{L}] = \hat{L}\rho\hat{L}^\dagger - \{\hat{L}^\dagger\hat{L}, \rho\}/2$. We consider dissipation corresponding to cavity loss $\hat{L}_c = \sqrt{2\kappa}\hat{a}$, exciton loss $\hat{L}_{\text{ex},j} = \sqrt{\Gamma_{\text{ex}}}\hat{X}_j$, and phonon loss $\hat{L}_{\text{b},j} = \sqrt{\Gamma_{\text{phon}}}\hat{b}_j$. The symbol definitions and values used are included in Table 2.

The cavity and pump frequencies are chosen to be close to that of the exciton, and so we move into a frame rotating at the mean pump frequency $\bar{\omega}$ and disregard counter-rotating terms. While these frequencies are close to resonance, we note that in the results shown below, we will work in a regime where the two-photon process (i.e., the effective photon-phonon coupling) are closer to resonance than the one-photon process of exciton absorption. We choose this regime in order to keep the exciton population and heating effects as small as possible. As noted above, the exciton population cannot necessarily be made sufficiently small to ignore, but keeping it as small as attainable leads to a simpler picture. Following the rotating-wave approximation, we then implement a mean-field ansatz (details given in Methods) and obtain coupled equations for the rescaled cavity field $\alpha = \langle \hat{a} \rangle / \sqrt{N}$ and the density matrix of a single site p :

$$\partial_t \alpha = -i \left[-\Delta_c \alpha + g_0 \sqrt{N} \text{Tr}(\rho \hat{X}) \right] - \kappa \alpha; \quad (2)$$

Fig. 1 | Proposed experiment schematic and energy level diagram. **a** Schematic of the scenario considered. Two pumps (red and blue beams) drive a two-dimensional material coupled to an optical cavity mode (pink). **b** Energy-level diagram showing the two electronic states, vibrational sub-bands, couplings of the exciton to the pumps (red, blue) and cavity (magenta), and detunings, as defined in Table 1.

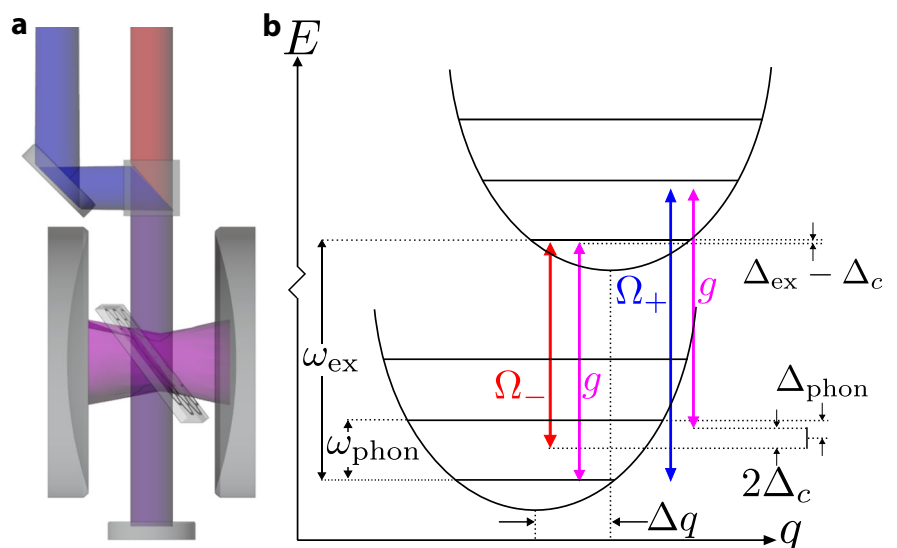


Table 1 | Frequencies and detunings used in the transverse pumping model

Symbol	Definition	Meaning
$\bar{\omega}$	$\equiv \frac{1}{2}(\omega_+ + \omega_-)$	Mean pump frequency
Δ_{ex}	$\equiv \bar{\omega} - \omega_{\text{ex}}$	Exciton detuning
Δ_c	$\equiv \bar{\omega} - \omega_c$	Cavity detuning
$\Delta\omega$	$\equiv \frac{1}{2}(\omega_+ - \omega_-)$	Half of the splitting of pump frequencies
Δ_{phon}	$\equiv \Delta\omega - \omega_{\text{phon}}$	Detuning from $\Delta\omega$ to the phonon energy
Δ_{ex}^D	$\equiv \Delta_{\text{ex}} + (\Delta q)^2 \omega_{\text{phon}}$	Dressed exciton detuning
Δ_c^D	$\equiv \Delta_c - \frac{g_0^2 N}{\Delta_{\text{ex}} - \Delta_c}$	Dressed cavity detuning

$$\partial_t \rho = -i[\hat{H}_{\text{mat}}^{\text{MF}}, \rho] + \mathcal{D}[\sqrt{\Gamma_{\text{ex}}}\hat{X}] + \mathcal{D}[\sqrt{\Gamma_{\text{phon}}}\hat{b}]. \quad (3)$$

The mean-field material Hamiltonian $\hat{H}_{\text{mat}}^{\text{MF}}$ is

$$\begin{aligned} \hat{H}_{\text{mat}}^{\text{MF}} = & -\Delta_{\text{ex}}\hat{X}^\dagger\hat{X} + \omega_{\text{phon}}\left[\hat{b}^\dagger\hat{b} + \Delta q\left(\hat{b} + \hat{b}^\dagger\right)\hat{X}^\dagger\hat{X}\right] \\ & + \left[\left(\Omega_+ \frac{e^{-i\Delta\omega t}}{2} + \Omega_- \frac{e^{+i\Delta\omega t}}{2} + g_0\sqrt{N}\alpha\right)\hat{X}^\dagger + \text{H.c.}\right], \end{aligned} \quad (4)$$

where the detunings are given in Table 1. The mean-field ansatz is appropriate in the limit we consider, where there is a very large number of emitters N coupled to a single photon mode; mean-field theory has been proven exact for open Dicke models in the limit of infinite N ³⁸.

There remains explicit time dependence in the exciton drive at frequency $\Delta\omega$. As noted above, and can be seen in Fig. 1(b), this detuning is included so that there can be resonant processes for scattering a phonon into a photon, as well as resonant processes to create a phonon-photon pair. Such resonance occurs if $\Delta_{\text{phon}} \equiv \Delta\omega - \omega_{\text{phon}} = 0$. In most prior work on Raman-driving schemes, the exciton state would be eliminated, and the above resonant processes would then lead to an effective coupling of the type $(\hat{a} + \hat{a}^\dagger)(\hat{b}_j + \hat{b}_j^\dagger)$. This adiabatic elimination is not so clearly justified in this work, as we cannot necessarily operate in a parameter regime where the exciton detuning is larger than all other detunings—we discuss relevant parameter values in the next subsection.

In the following, we will solve this periodically driven and dissipative mean-field model. We will first consider solutions with time dependence commensurate with the drive frequency; these are analogous to the steady-state solutions found for a time-independent model. We will refer to such solutions as *stationary states*. As discussed further below, not all solutions take this form—analogue to limit cycles in a time independent problem, one can also sometimes find solutions incommensurate with the drive frequency. When considering the stationary state, we can avoid needing to perform direct time evolution of Eqs. (2) and (3), and can instead directly find the stationary states by looking for solutions in the form of a Fourier series: $\alpha(t) = \sum_n \alpha_{ss,n} e^{in\Delta\omega t}$, and $\rho(t) = \sum_n \rho_{ss,n} e^{in\Delta\omega t}$, where the sum over n is truncated at some maximum $|n|$. See Methods for details on the solving process for the Fourier coefficients. Figure 2 shows that this procedure well matches the results of direct time evolution. This process can only find solutions analogous to steady states of a time-independent driven system; direct time evolution must be used to find analogues to limit cycles, which may have time scales far longer than the periodic driving.

The stationary-state solver always finds a normal state with $\alpha_{ss,0} = 0$, i.e., one with zero steady population of the cavity. However, because the pumps are still interacting with excitons and excitons continue to scatter into the cavity, the nonzero-frequency components of α_{ss} in the normal state do not vanish—specifically terms with odd n are nonzero, as seen in Fig. 2. While this normal state always exists, it is not always stable. To determine this, we linearise the master equation about one of its stationary states. This results in an equation in Floquet form. The Floquet exponents found from this

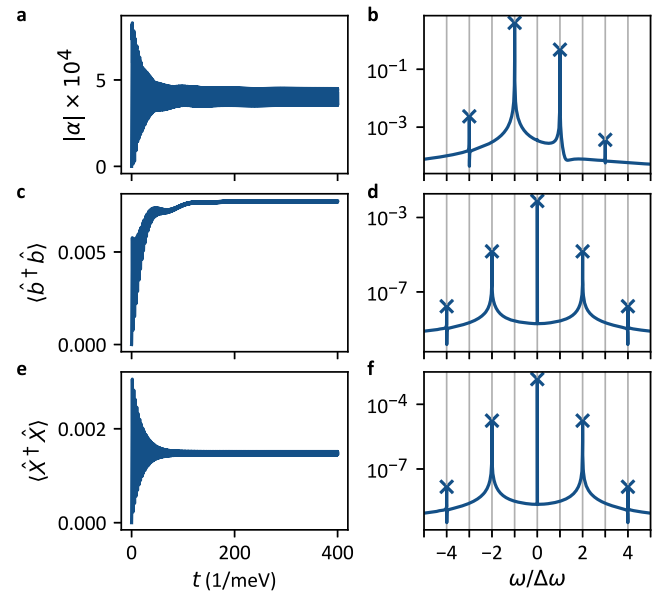


Fig. 2 | Comparison between direct time evolution of Eqs. (2) and (3) and the stationary-state solution in the form of a Fourier series. Shown are three different observables: **a, b** Photon field; **c, d** phonon number; and **e, f** exciton number. Left plots show the time evolution. Right plots show, on a logarithmic scale, the magnitudes of the discrete Fourier transforms of the late-time evolution shown as lines from 900 to 1000/meV compared to the Fourier components present in the stationary-state solution shown as crosses. For the time-evolution results, each peak is broadened due to the finite duration of the simulation. Parameter values are $\Delta_c = -0.25$ meV and $\Omega_+ = 0.2$ meV, along with those listed in Table 2; the system is initialised in the vacuum state.

equation then determine the stability of the stationary state: If any Floquet exponent has real part greater than zero, then the state is unstable. For our time-dependent system, these Floquet exponents play the role that linear-stability eigenvalues played in previous work³⁹. We therefore refer to these Floquet exponents as eigenvalues in what follows; see Methods for further details. Regimes where the normal state with $\alpha_{ss,0} = 0$ is stable we term the *normal phase*. Regimes where the normal state is unstable are discussed below, as we find multiple other states that may arise.

Low-energy effective model

Before presenting results found directly from the stationary state and stability analysis of the full photon-exciton-phonon model discussed in the previous section, we introduce an effective model that provides a qualitative understanding of the results. This effective model results from adiabatic elimination of the exciton states, leading to an effective photon-phonon model, as discussed in the Supplementary Information⁴⁰. It takes the form $\hat{H}_{\text{eff}} = -\Delta_c^D \hat{a}^\dagger \hat{a} + \sum_{j=1}^N \hat{H}_{\text{eff},j}$, with

$$\hat{H}_{\text{eff},j} = \sum_{j=1}^N -\Delta_{\text{phon}} \hat{b}_j^\dagger \hat{b}_j + \left(\lambda_+ \hat{a} \hat{b}_j + \lambda_- \hat{a} \hat{b}_j^\dagger + \text{H.c.} \right). \quad (5)$$

This effective model is similar to the generalised Dicke model⁴, with distinct co- and counter-rotating couplings (λ_+ , respectively). However, the effective model in Eq. (5) differs from the Dicke model in that it replaces two-level systems by bosons (phonons); this would change the behaviour of such a model above threshold, but does not modify the location of the threshold. The phase diagram of the generalised Dicke model has been studied in several places^{41–45}. We will refer to these works, particularly⁴², to interpret features of the phase diagrams we find in the following.

We can derive approximate perturbative forms for the couplings $\lambda_{\pm}$ and the dressed-cavity detuning Δ_c^D appearing in this model in the limit

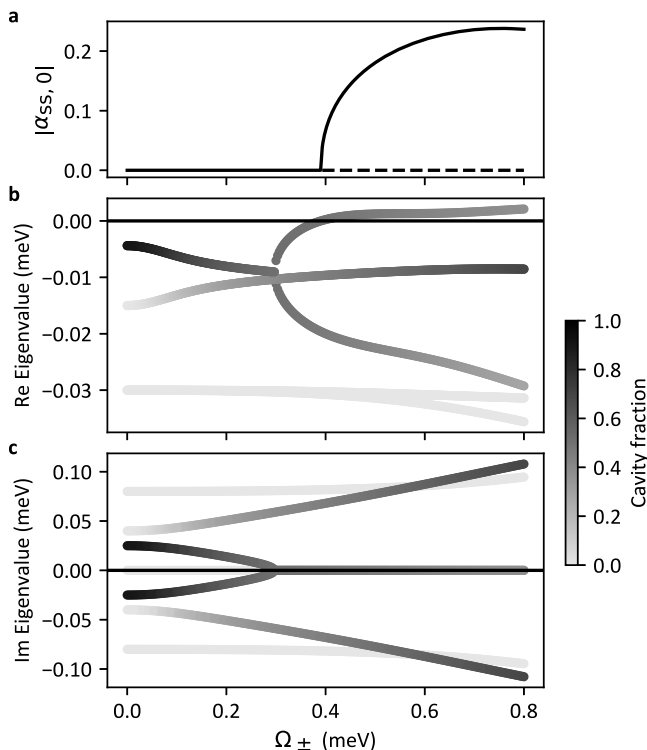


Fig. 3 | Pitchfork bifurcation leading to a superradiant state. **a** Zero-frequency component of the stationary-state cavity amplitude $\alpha_{ss,0}$, showing the emergence of the superradiant phase above the bifurcation at $\Omega_{\pm} \approx 0.4$ meV. **b**, **c** Real and imaginary parts of the eigenvalues of the Floquet system linearised about the normal state. Only the 7 nontrivial eigenvalues with the greatest real parts are shown. Shading indicates the fraction of cavity mode in the corresponding eigenvectors, such that darker shading indicates more photonic modes. Parameters are in Table 2, along with $\Delta_c = -0.25$ meV.

where the phonon energy ω_{phon} is close to $\Delta\omega$ and all detunings are much smaller than this scale: i.e., for $\Delta_{\text{ex}}^D, \Delta_c, \Delta_{\text{phon}} \ll \omega_{\text{phon}} \approx \Delta\omega$. Here we have introduced the dressed exciton detuning $\Delta_{\text{ex}}^D = \Delta_{\text{ex}} + \omega_{\text{phon}} \Delta q^2$, accounting for polaron dressing of the exciton. That is, Δ_{ex}^D corresponds to detuning between the pump and the polaron energy, where the polaron energy is the lowest energy attainable in the one-exciton manifold, corresponding to minimising the Hamiltonian $\omega_{\text{phon}}[\hat{b}^\dagger \hat{b} + \Delta q(\hat{b}^\dagger + \hat{b})\hat{X}^\dagger \hat{X}]$. In the limit where all detunings are much smaller than ω_{phon} , we find:

$$\lambda_{\pm} = \frac{1}{4} \Omega_{\pm} g_0 \Delta q \left(\frac{1}{\Delta_{\text{ex}}^D \pm \Delta_{\text{phon}}} + \frac{1}{\Delta_{\text{ex}}^D - \Delta_c} \right), \quad (6)$$

along with $\Delta_c^D = \Delta_c + g_0^2 N / (\Delta_{\text{ex}}^D - \Delta_c)$. This dressed cavity detuning accounts for hybridisation of photons with excitons. As well as modifying this detuning, this hybridisation also modifies loss terms, so that cavity loss is increased: $\kappa \rightarrow \kappa^D = \kappa + \frac{1}{2} g_0^2 N \Gamma_{\text{ex}} / (\Delta_{\text{ex}}^D - \Delta_c)^2$. In⁴⁰, we provide an alternative numerical approach to give non-perturbative estimates of these dressed energies and decay rates.

While the model above does not provide quantitatively accurate results, it does indicate what conditions might favour strong coupling between phonons and photons, and thus photon-polariton condensation. From the form of Eq. (6), there are two possible conditions that lead to large values of $\lambda_{\pm}$, corresponding to either of the two denominators approaching zero. These two cases correspond to the exciton being in resonance with the cavity, or to a vibrational sideband of the exciton being in resonance with one of the pumps. In both cases, this can lead to a large population of excitons, thus invalidating the adiabatic elimination, and furthermore causing heating due to exciton decay. The ideal scenario is to be very close to

the two-photon resonance (i.e., where the Raman process resonantly creates or destroys a phonon), while avoiding the one-photon resonance that leads to large exciton populations. We also note that Eq. (6) suggests that the denominators can be made arbitrarily small, leading to arbitrarily large coupling. However, exciton and phonon decay rates introduce cutoffs on these denominators. This consideration motivates the values of the control parameters $\Delta_{\text{ex}}^D, \Delta_c, \Delta_{\text{phon}}$ used in this work, in that we generally probe the regimes $|\Delta_{\text{ex}}^D| \gtrsim \Gamma_{\text{ex}}$ and $|\Delta_{\text{phon}}| \gtrsim \Gamma_{\text{phon}}$.

Superradiant phase transition

Considering the full photon-exciton-phonon model of Sec. II A, we next discuss the ways in which the system may transition into a superradiant state (i.e., to a phonon-polariton condensate). As discussed above, the $\alpha_{ss,0} = 0$ stationary state may go from stable to unstable as indicated by the eigenvalues of the Floquet problem. Such instability can arise as one increases the driving strength. We discuss here two scenarios of what may happen at this instability, associated with continuous and discontinuous phase transitions.

In the continuous case, the instability of the normal state coincides with the appearance of a new stable stationary state in which $\alpha_{ss,0} \neq 0$. Such states correspond to a macroscopic population of the cavity that spontaneously breaks the symmetry of the model. In the original frame, $\alpha_{ss,0}$ corresponds to cavity light at the mean pump frequency $\bar{\omega}$: The appearance of light at this mean frequency is understood as the signature of a superradiant state^{4,36}. Stationary states with $\alpha_{ss,0} \neq 0$ must come in equal and opposite pairs in the complex plane because the master equation has a (weak^{46,47}) symmetry under the transformation $(\alpha, \tilde{X}) \leftrightarrow (-\alpha, -\tilde{X})$. As such, the appearance of nonzero $\alpha_{ss,0}$ corresponds to spontaneous symmetry breaking. An illustration of this scenario is shown in Fig. 3. Zooming in near the transition, we see it occurs when a single, real eigenvalue goes from negative to positive, corresponding to a pitchfork-bifurcation instability⁴⁸. This corresponds to the standard continuous phase transition to the Hepp-Lieb-Dicke superradiant phase⁸.

The critical pumping strength may be estimated using the low-energy effective model introduced above. For effective couplings that are nearly equal, $|\lambda_- - \lambda_+| \ll \lambda_- + \lambda_+$, the transition occurs when⁸

$$\lambda_- \lambda_+ N = \frac{1}{4} \left(\frac{(\Delta_c^D)^2 + (\kappa^D)^2}{\Delta_c^D} \right) \left(\frac{\Delta_{\text{phon}}^2 + \Gamma_{\text{phon}}^2/4}{\Delta_{\text{phon}}} \right). \quad (7)$$

The right-hand side of this expression reduces to the more familiar $\Delta_c^D \Delta_{\text{phon}} / 4$ if loss is ignored. However, including effects of loss is crucial for understanding the minimal threshold pump power attainable, as we discuss further in Sec. II E.

A notable feature of the low-energy effective model is the replacement of bare cavity loss by the effective loss rate κ^D . This effect can be seen directly from Fig. 3b. The intercepts of each line with the vertical axis indicate the linewidths of the effective modes in the absence of pumping, i.e., when $\lambda_{\pm} = 0$. One may see that the darkest line, corresponding to the cavity mode, has an intercept much below $-\kappa$, where $\kappa = 6 \times 10^{-4}$ meV. We previously gave the perturbative expression for the dressed cavity linewidth as $\kappa^D = \kappa + \frac{1}{2} g_0^2 N \Gamma_{\text{ex}} / (\Delta_{\text{ex}}^D - \Delta_c)^2$. For the parameters of Fig. 3(b), this yields $\kappa^D \approx 0.009$ meV, while the intercept in the plot is -0.004 meV. The discrepancy is due to the large value of the parameter that controls the perturbation expansion, $g_0 \sqrt{N} / (\Delta_{\text{ex}}^D - \Delta_c) \approx 0.5$. One may note that the expansion overestimates the broadening because it disregards the eventual saturation of the dressed cavity linewidth at very large coupling $g_0 \sqrt{N}$: In the large-coupling limit, the relevant mode becomes an equal superposition of cavity and exciton, with a corresponding linewidth $\kappa^D = \frac{1}{2} \kappa + \frac{1}{4} \Gamma_{\text{ex}}$.

In other parameter regimes, the transition to the superradiant state can be discontinuous. An example of this is shown in Fig. 4, where a stable superradiant state emerges from a saddle-node bifurcation at nonzero $\alpha_{ss,0}$ at a particular driving strength Ω . There is a range of driving strengths above this for which the mean-field theory predicts bistability. At the upper limit of

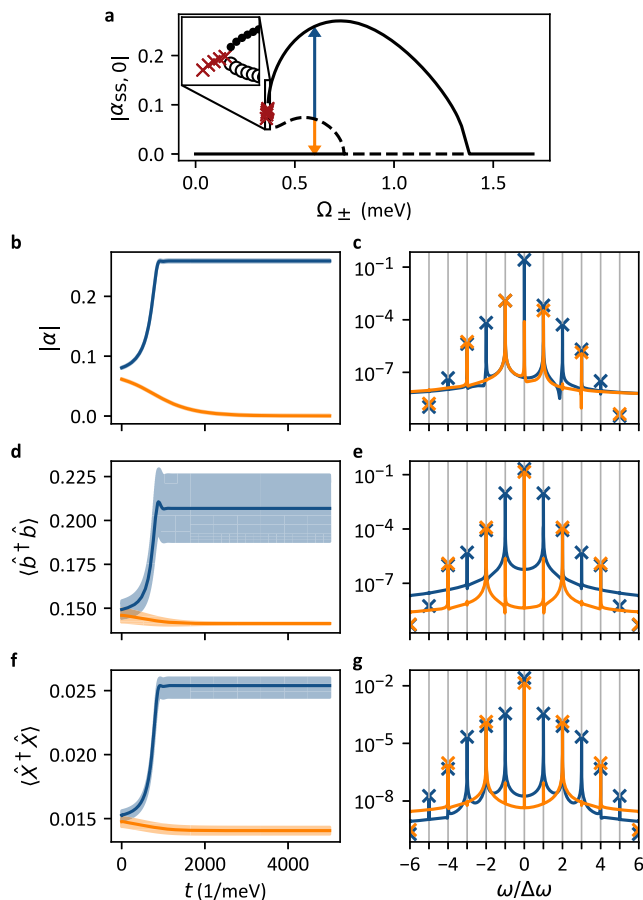


Fig. 4 | Superradiant bistability. The system is evolved from two initial states labelled with blue and orange throughout. The upper, blue lines approach the stable superradiant phase, while the lower, orange lines approach the stable normal phase, indicating bistability. In **a** the stable stationary states are joined by solid lines, and the unstable states by dashed lines. The red crosses are spurious stationary states found by the solver. Time evolution plots and their Fourier components obtained via both time evolution (lines) and the stationary state solver (crosses) for: (**b**, **c**), the cavity amplitude; (**d**, **e**), the phonon number; and (**f**, **g**), the exciton number. The solid lines on the left are moving averages taken over 2 periods. Fourier components are taken from the time evolution between 4000 to 5000/meV. Even at these late times, the cavity amplitude's zero-frequency component and the phonon and exciton number's $\pm 1\Delta\omega$ frequency components have not fully decayed. Parameter values are in Table 2, along with $\Delta_c = -0.24$ meV and $\Omega_{\pm} = 0.6$ meV.

the bistable region, the normal phase goes unstable at a subcritical rather than a supercritical pitchfork bifurcation⁴⁸. This case allows for hysteresis, and it illustrates an unusual first-order superradiant transition^{49,50}, as with other models where mean-field theory supports bistability, beyond-mean-field treatments are expected to predict a unique discontinuous phase transition, determined by the switching rates between the $\alpha_{ss,0} = 0$ and $\alpha_{ss,0} \neq 0$ solutions. Although this transition is discontinuous, we note it still involves spontaneous symmetry breaking to pick the sign of $\alpha_{ss,0}$.

In addition to exhibiting a discontinuous transition, Fig. 4 also shows another notable feature: A transition back to the normal state at larger pump strengths. Similar behaviour has been discussed in other lasing or superradiance models, e.g.,⁵¹—strong pumping can cause an increase of effective linewidth, i.e., a form of power broadening⁵². We note that such an effect is often lacking in approaches that have adiabatically eliminated intermediate states involved in the pumping.

Lasing transition

The emergence of a superradiant state is not the only manner in which the normal phase can go unstable. There is also a scenario where two complex-

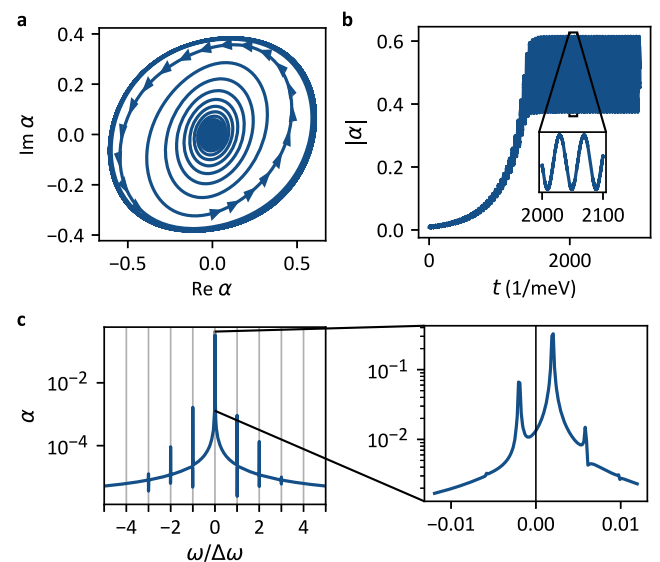


Fig. 5 | Lasing. The system is initialised close to the unstable stationary state near the vacuum. **a** Real and imaginary parts of the cavity amplitude α . **b** Time evolution of α , indicating the slow approach to the limit cycle and its associated long time scale. **c** Fourier transform of the late time evolution, from $t = 2000$ to 3000 /meV. The inset shows behaviour near zero frequency, indicating that the low-frequency peak is detuned away from zero frequency. Parameter values are in Table 2, along with $\Delta_c = -0.08$ meV and $\Omega_{\pm} = 1$ meV.

conjugate eigenvalues simultaneously cross the imaginary axis. This corresponds to a Hopf bifurcation⁴⁸ and is expected to lead to a limit cycle. Indeed, as seen in Fig. 5, we find that in this case the cavity amplitude α spirals out from the origin and undergoes large-scale oscillations on a timescale that is much longer than the period of the drive, $1/\Delta\omega$. That is, the time dependence of this solution is not commensurate with the drive. Near to the transition, the frequency of these slow oscillations matches the imaginary part of eigenvalue, as is typical for a Hopf bifurcation. Because the stationary-state solver assumes that the final frequency components are integer multiples of $\Delta\omega$, it cannot find these incommensurate solutions. As such, we use direct integration of the master equation to check the stability of the limit cycle.

We denote this behaviour the *lasing* phase, in analogy to the phases found in the generalised Dicke model. After adiabatic elimination of excitons, the generalised Dicke model (as found in Sec. II B) is time-independent in the frame of the average pump frequency. One can distinguish time-independent (superradiant) and time-dependent (lasing) states in such a case. Reference⁴² showed that there are two different parameter regimes where the generalised Dicke model exhibits lasing. The first is the typical lasing regime, where an incoherent drive leads to inversion of the emitters, causing gain for the cavity mode. However, the model of Sec. II B has no incoherent drive, only decay, and the emitters in Eq. (6) are bosons, not two-level systems. As such, inversion of emitters is not relevant for the case we consider here. The second lasing state has a different character. This state is found either when $\lambda_+ \gg \lambda_-$, or when the sum of the cavity and emitter frequencies approach zero. In either of these cases, the dominant matter-light coupling is one that creates a photon and *excites* an emitter, the counter-rotating coupling. This then leads to a state that has been referred to as “counter-lasing”^{44,45}.

In the case shown in Fig. 5 (and also discussed further below), our time-dependent phase can be ascribed to counter-lasing driven by near-resonance of the process that creates a photon and excites an emitter; see, e.g., Fig. 8 of Ref. 42. Such resonance would occur if $\Delta_c^D = -\Delta_{\text{phon}}$. Even though the bare detuning Δ_c is negative in Fig. 5, corresponding to red detuning, the shift due to the exciton-cavity coupling can push Δ_c^D to become positive, corresponding to effective blue detuning. In principle,

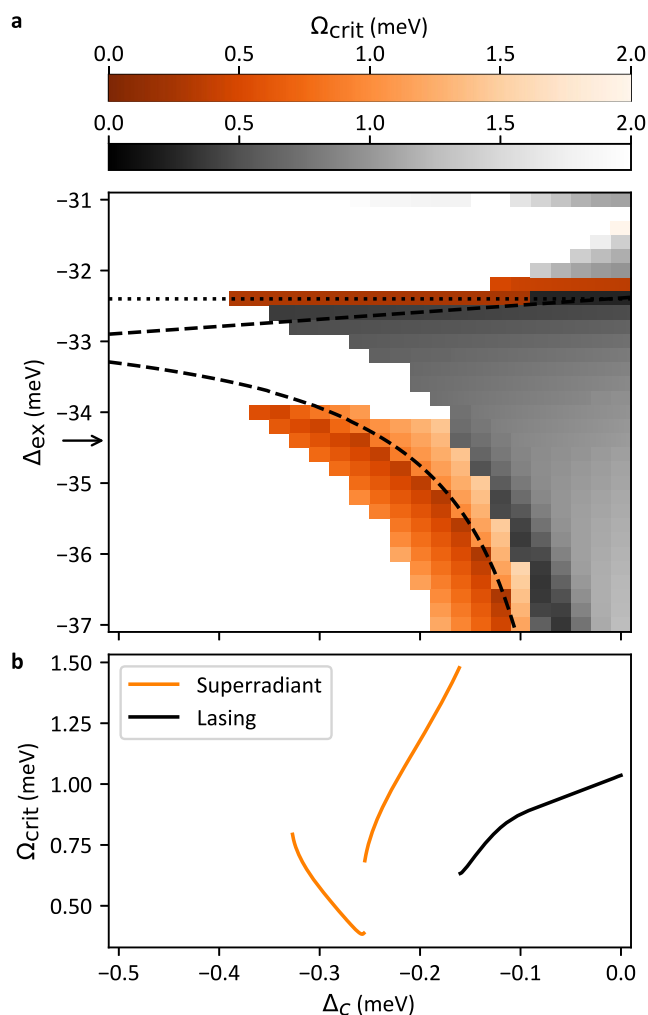


Fig. 6 | Phase diagram indicating the minimum pump strength at which the normal phase goes unstable. The two colours indicate whether one eigenvalue goes unstable (orange) corresponding to the superradiant transition, or two eigenvalues go unstable (black) corresponding to lasing. Parameters are in Table 2, along with $\Omega_- = \Omega_+$. **a** The dotted line indicates where $\Delta_{\text{ex}}^D = 0$, i.e., where Δ_{ex} crosses the polaron energy $-\omega_{\text{phon}}\Delta q^2 = -32.4$ meV. The dashed lines separate the regions where the cavity eigenvalue at zero pump, i.e., at $\Omega_{\pm} = 0$, has an imaginary part that is positive (lower right region) versus one that is negative (lower left and upper regions). These regions correspond to the cavity being effectively blue-detuned and red-detuned, respectively. The arrow indicates the value of Δ_{ex} for the slice in **b**. **b** Horizontal slice of the phase diagram of (a) at the location indicated by the arrow, $\Delta_{\text{ex}} = -34.4$ meV.

counter-lasing may also occur in our model due to $\lambda_+ \gg \lambda_-$, but this requires larger Δ_{phon} and takes the system out of the small-detuning regime used to derive $\lambda_{\pm}$.

Phase Diagram

Having discussed the forms of instability that can arise, we now discuss the phase diagram, indicating which forms of instability occur for certain parameters. A key practical question is what critical pump strength is required to reach either the superradiant or lasing instability. Figure 6 shows this diagram, mapping out the critical pump strength at which the normal phase goes unstable due to a single eigenvalue (superradiant state) or a pair of complex-conjugate eigenvalues (lasing). Dark shading indicates where a transition can occur at low pump power. White regions indicate parameters for which no instability of the normal state is found.

Experimentally exploring this full phase diagram requires tuning both the mean pump frequency $\bar{\omega}$ and the cavity frequency. Changing only the

mean pump frequency corresponds to diagonal traces through this figure from lower left to upper right. The superradiant and lasing phases appear continuously connected in the space of Δ_c and Δ_{ex} in some regimes, but for other regimes they are separated. Details of this structure can be seen in the cross section in Fig. 6(b). A jump in the critical pump strength within the superradiant phase is also exhibited. This is associated with switching between the continuous and discontinuous phase transitions. To see how this scenario evolves in more detail, Supplementary Note 2⁴⁰ shows how eigenvalues coming close to but not crossing the imaginary axis can result in jumps in the transition pumping strength.

A notable feature of this figure is the change in behaviour as the exciton detuning Δ_{ex} is tuned through $-\omega_{\text{phon}}\Delta q^2$ as indicated by the dotted line. This corresponds to Δ_{ex}^D crossing zero, or equivalently, to tuning the mean pump frequency through the polaron energy. At this detuning we see a narrow sliver of superradiant state. One may note that the low-energy effective model does not apply under this condition because resonance results in stationary states that have high-exciton population even at low pump strength. A second consequence of small Δ_{ex}^D is a very large shift to the dressed photon frequency; i.e., Δ_c^D is large and positive just below $\Delta_{\text{ex}}^D = 0$, and large and negative just above this line. This in turn explains why the superradiant state ceases for $\Delta_{\text{ex}} > -\omega_{\text{phon}}\Delta q^2$.

A second important feature occurs when Δ_c^D changes sign, as shown by the dashed curves in Fig. 6. As discussed in the previous section, counter-lasing occurs in general for positive Δ_c^D , and in particular when $\Delta_c^D \simeq -\Delta_{\text{phon}}$. In contrast, the superradiant state occurs for negative Δ_c^D . Since Fig. 6 is plotted at fixed $\Delta\omega$ and thus fixed Δ_{phon} , the switch from the superradiant phase to counter-lasing is driven by changing Δ_c^D .

As can be understood from the low-energy effective model (i.e., Eq. (6) and Eq. (7)), the points of particularly low threshold are close to points where the transition ceases to exist. This corresponds to the standard observation that with dissipation, the lowest threshold occurs when $\Delta_c^D = \kappa^D$, while the transition vanishes at $\Delta_c^D = 0$. In Fig. 6(a), the dashed line corresponding to $\Delta_c^D = 0$ does not exactly match the point where the superradiant transition ceases; this is because the estimate of Δ_c^D used here neglects effects of the AC Stark shift of the exciton due to the finite pump strength $\Omega_{\pm}$. In contrast, calculations in Ref. 40 performed for the low-energy effective model (i.e., neglecting this AC Stark shift) do show the expected agreement.

Discussion

In the results above we have shown that, considering a standard model of Holstein coupling between excitons and phonons, one can realise a Raman-driving scheme to couple Raman-active phonons to light. One can thus realise a phonon-polariton condensate that is indicated by a steep jump in cavity photon emission. Furthermore, below threshold all emitted light is at the frequencies of the pumps; above threshold, light will come out at the average of these frequencies (superradiant phase) or at an incommensurate frequency (lasing phase). The corresponding changes to the phonon mode frequencies can be further verified with in situ Raman spectroscopy.

While an effective model that adiabatically eliminates the excitons can qualitatively describe the phases, we found that this adiabatic elimination is not quantitatively accurate for the chosen parameters relevant to TMDs. In principle, for materials with very large band gaps and thus large exciton energies, such adiabatic elimination may be adequate. However, for the materials we consider, one must use a more sophisticated method that retains the exciton states and thus requires a treatment of a periodically driven system. Our results are written in terms of the driving strength $\Omega_{\pm}$ appearing in Eq. (1). Here we discuss the practicality of attaining these values in terms of required laser power. We also discuss design principles for choices of materials as well as open questions arising from this work.

To estimate the laser power required to reach the superradiant transition, we make several simplifications to obtain a closed-form expression. First, we set the detunings Δ_{ex}^D and Δ_{phon} to half the exciton and phonon linewidths, respectively. Second, we assume we work in the regime of large exciton-cavity coupling; in this regime, as discussed above, the dressed-

cavity linewidth $\kappa^D \simeq \Gamma_{\text{ex}}/4$, a linear scaling with exciton linewidth. (We discuss the other possible regimes in Ref. 40.) Third, we set the denominators in the effective couplings Eq. (6) to Γ_{ex} because Δ_{ex}^D is the largest energy scale in the denominators and is assumed to be dressed by exciton decay of an amount between $\frac{1}{2}\Gamma_{\text{ex}}$ and Γ_{ex} . Finally, we choose equal pumping, so $\Omega_{\pm} = \Omega$. With these choices, the threshold condition becomes $(\Omega_{\text{crit}} g_0 \Delta q)^2 N = \frac{1}{2} \Gamma_{\text{ex}}^3 \Gamma_{\text{phon}}$.

To convert this from a critical value of Ω to a critical laser power, we must relate the light-matter coupling parameters in our model to electric field strengths of the cavity and pump light. The exciton-light couplings can be written as $\Omega\sqrt{N} = e_{\text{ex}}\sqrt{AE_p}$ and $g_0\sqrt{N} = e_{\text{ex}}\sqrt{AE_c}$ for pump and cavity electric fields $E_{p,c}$, material area A , and exciton matrix element e_{ex} ; see⁵³ and⁴⁰ for details. We write the number of emitters as $N = nA$ for emitter density n . Assuming the pump is a Gaussian beam impinging onto the material area, its power is $P = \frac{1}{4}\epsilon_0 E_p^2 A c$. The threshold pump power is then:

$$P_{\text{crit}} = \frac{\Gamma_{\text{ex}}^3 \Gamma_{\text{phon}} n \epsilon_0 c}{8 e_{\text{ex}}^4 \Delta q^2 E_c^2}. \quad (8)$$

We see from this expression that the critical pump power depends separately on the density of emitters n and the matrix element e_{ex} , and not just upon the combination $e_{\text{ex}}/\sqrt{n}$ that controls the coupling strength g_0 . Since the materials we wish to consider do not actually consist of a set of localised emitters, a choice must be made to give separate estimates of n and e_{ex} . In the following we will set the emitter density to the inverse unit cell area—alternative choices are the Mott density and inverse exciton area⁵⁴.

We take MoSe₂ as our model material, so we set $\Gamma_{\text{ex}} = 1$ meV, $\Gamma_{\text{phon}} = 0.05$ meV, $e_{\text{ex}} = 0.23e$, $\Delta q^2 = 1$, and $n^{-1} = \sqrt{3}a_0^2/2$ for a lattice spacing of $a_0 = 3.3$ Å^{26,29,30,53}. We set the cavity waist to $w_0 = 30$ μm and quantise its electric field over cavity length $L = 1$ cm with a Gaussian mode profile; note that a cavity this long allows for transverse pumping and cryogenic cooling of a sample in situ. This yields a threshold pump power of $P_{\text{crit}} = 30$ W. While this is too great to be immediately feasible, we will remark later on how a multimode cavity may bring this down to a practical regime. Note that the dependence of the critical power on e_{ex}^4 and Γ_{ex}^3 mean small changes in estimates of these parameters have large effects on the critical power.

As mentioned before, the model we discuss is not specific to TMDs and can be applied to other materials. This prompts the question of which material properties would be best-suited to realising phonon-polariton condensation. Based on the results above, an estimate of this can be found by considering how to maximise the effective coupling defined in Eq. (6) or minimise the threshold power of Eq. (8). These expressions highlight the beneficial effects of the exciton matrix element and the Huang-Rhys parameter $S = \Delta q^2$ defining the exciton-phonon coupling, as well as the adverse effect of the exciton linewidth.

To conclude this discussion, we briefly mention some topics that could be addressed in future work.

Multimode cavities

This paper considers a single-mode cavity for simplicity, but experiments using a degenerate confocal cavity⁹ have been realised for ultracold atoms. In such confocal cavities, one can obtain a field enhancement^{10,55} by constructive interference between different degenerate modes building a synthetic cavity mode localised to have perfect overlap with the cloud of atoms. Such an approach can also be used in this solid-state context to maximise overlap with whichever is smaller, the sample or the pump spot. Such an approach thus allows one to further enhance the cavity-exciton coupling. In particular, a multimode cavity may reduce the threshold laser power required. To continue the calculation from before, if one considers a pump beam waist of $w_p = 5$ μm, then the confocal enhancement would be $(w_0/w_p)^2 = 36$, reducing the critical pump threshold to below 1 W. A more sophisticated treatment of multiple cavity modes may however need to go beyond the mean-field ansatz. Mean-field approaches may remain valid when the number of emitters is larger than the number of relevant cavity modes⁵⁶; exploring this question is the subject of future work.

Temperature

In our calculations above, we implicitly assumed a low-temperature state, in which there is no population of phonons in the initial state. Such an approach is reasonable when considering experiments at cryogenic temperatures, i.e., when the temperature is much smaller than the phonon energy. Cryogenic cooling to this low-temperature state should also allow one to minimise heating effects from the continuous-wave pumping. Extending the calculations here to consider materials at higher temperature (e.g., room temperature) is an open question. In particular, at higher temperatures it may be important to distinguish between the exciton and phonon linewidths due to decay versus dephasing. As has been explored in simple models⁴², dephasing and decay have different effects on the threshold for condensation.

Effects on electronic states

A key question for future work is to assess the effect of phonon polariton condensation on electronic and transport properties, following methods such as those in Refs. 2,3,20. A related question is to explore whether other states can be realised by the Raman pumping scheme in this work, such as more complex limit cycles or chaotic phases.

Methods

Transformation to mean-field Hamiltonian

To transform the original Hamiltonian, Eq. (1), into the frame rotating at $\bar{\omega}$, we use the unitary transform:

$$\hat{U}_{\text{rot}} = \exp \left[i \bar{\omega} t \left(\hat{a}^\dagger \hat{a} + \sum_{j=1}^N \hat{X}_j^\dagger \hat{X}_j \right) \right]. \quad (9)$$

To derive the model in Eqs. (2)–(4), we then disregard terms rotating at $2\bar{\omega}$ and $\bar{\omega} + \omega_{\pm}$. We then make a mean-field approximation, equivalent to assuming the density matrix factorises into terms for the cavity and for each site. With this approximation, the equation of motion for the cavity becomes that of a coherently driven damped harmonic oscillator, and so its behaviour is captured by considering the evolution of the coherent state amplitude:

$$\alpha \equiv \frac{\langle \hat{a} \rangle}{\sqrt{N}}. \quad (10)$$

For the matter component, we assume all sites are equivalent, leading to the site-independent model in Eq. (4).

Fourier analysis and linear stability

The Fourier series ansatz for stationary states transforms the system of nonlinear differential equations for (α, ρ) into a system of nonlinear algebraic equations for $(\alpha_{\text{ss},n}, \rho_{\text{ss},n})$. All nonlinearity here arises from the dependence on $\alpha_{\text{ss},n}$. As such, at fixed $\alpha_{\text{ss},n}$, the problem simplifies into a linear homogeneous system for $\rho_{\text{ss},n}$. The trace constraint $\text{Tr}(\rho_{\text{ss},n \neq 0}) = 0$ is already enforced by the system, but we must manually add the normalisation constraint $\text{Tr}(\rho_{\text{ss},0}) = 1$ to one of the rows. We then solve this exactly determined system for $\rho_{\text{ss},n}$ via sparse LU factorisation, and we feed the result back into the objective function for $\alpha_{\text{ss},n}$. This reduces the size of the nonlinear system from the full space of $(\alpha_{\text{ss},n}, \rho_{\text{ss},n})$ to the $2M + 1$ complex numbers $\alpha_{\text{ss},n}$, where M is the maximum $|n|$ retained in the sum; i.e., M is the cutoff on Fourier components. We also find the Jacobian of the objective function of $\alpha_{\text{ss},n}$ to accelerate the nonlinear root-finding. The Fourier cutoff M is typically set to 4, but it is occasionally set to 6 for detailed comparison to time evolution, as indicated in Fig. 4 and Table 2. We use the default settings in both the sparse LU factorisation and the root-finding using the Levenberg-Marquardt algorithm^{57,58}, as implemented in SciPy⁵⁹ following MINPACK⁶⁰.

The stationary-state solver returns a solution $(\alpha_{\text{ss}}(t), \rho_{\text{ss}}(t))$ with period $T = 2\pi/\Delta\omega$. To determine linear stability, we assume a small fluctuation about this stationary state: $\alpha(t) = \alpha_{\text{ss}}(t) + \delta\alpha(t)$ and $\rho(t) = \rho_{\text{ss}}(t) + \delta\rho(t)$, from which we obtain a system of differential equations for $(\delta\alpha(t), \delta\rho(t))$ that is linear but with explicit, periodic time dependence.

Table 2 | Full parameter list including values used in simulations

Symbol	Meaning	Value (meV)
ω_{phon}	Phonon energy	40
Δ_{ex}	Exciton detuning $\bar{\omega} - \omega_{\text{ex}}$	− 34.5, varies
Δ_{c}	Cavity detuning $\bar{\omega} - \omega_{\text{c}}$	varies
$\Delta\omega$	Detuning from the mean pump to either pump $ \bar{\omega} - \omega_{\pm} $	39.96
Δ_{phon}	Detuning from $\Delta\omega$ to the phonon energy $\Delta\omega - \omega_{\text{phon}}$	− 0.04
$\Omega_{\pm}$	Exciton-pump Rabi frequency	varies
$g_0\sqrt{N}$	Mean-field exciton-cavity coupling	1
Γ_{ex}	Exciton number decay rate	0.06
Γ_{phon}	Phonon number decay rate	0.03
κ	Mean-field cavity amplitude decay rate	6×10^{-4}
Symbol	Meaning	Value
Δq	Exciton-phonon coupling scaled by ω_{phon} ; Huang-Rhys parameter $S = \Delta q^2$	0.9
n_{ex}	Maximum exciton number	3
n_{phon}	Maximum phonon number	7
M	Fourier cutoff	4, 6

Writing this system as $\partial_t \vec{x} = \hat{A}(t)\vec{x}$ for $\vec{x} = (\delta\alpha, \delta\rho)$ and T -periodic $\hat{A}(t)$, we integrate to find the one-period evolution operator, also termed the monodromy matrix⁶¹:

$$\hat{\mathcal{M}} = T_{\leftarrow} \exp \left[\int_0^T \hat{A}(t) dt \right], \quad (11)$$

where $T_{\leftarrow}$ is the time-ordering symbol. We perform this integral by exponentiating and multiplying over discrete time steps of size $T/100$; we find this faster and more stable than any more sophisticated methods. The slow step is the subsequent diagonalisation of $\hat{\mathcal{M}}$, yielding eigenvalues $e^{\mu_k T}$ for Floquet multipliers μ_k . As mentioned before, these μ_k are called eigenvalues in this manuscript.

Direct time evolution of the system is performed using Netlib's zvode, as implemented in SciPy⁵⁹. We use the implicit Adams method at order 12, with rtol and atol parameters of 10^{-9} and 10^{-12} , respectively, and with a maximum number of steps of 1000.

Table 2 provides a summary of parameter values used in our numerical calculations. Throughout, QuTiP⁶² is used to create sparse matrices and to interpret density matrices, though the mean-field setup is not directly amenable to QuTiP methods.

Data availability

The data that support the findings of this study are available in the Harvard Dataverse Repository, <https://doi.org/10.7910/DVN/YGHFH2>.

Code availability

The code used for the current study is available in the Harvard Dataverse Repository, <https://doi.org/10.7910/DVN/YGHFH2>.

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Author contributions

B.L.L. and J.K. designed the research. A.N.B. performed the computations with input from J.K. All authors contributed to writing the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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