Superradiant Quantum Materials

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There is currently great interest in the strong coupling between the quantized photon field of a cavity and electronic or other degrees of freedom in materials. A major goal is the creation of novel collective states entangling photons with those degrees of freedom. Here we show that the cooperative effect between strong electron correlations in quantum materials and the long-range interactions induced by the photon field leads to the stabilization of coherent phases of light and matter. By studying a two-band model of interacting electrons coupled to a cavity field, we show that a phase characterized by the simultaneous condensation of excitons and photon superradiance can be realized, hence stabilizing and intertwining two collective phenomena which are rather elusive in the absence of this cooperative effect.

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Introduction.—Collective phenomena due to interactions between light and matter have become in recent years a major focus of interest spanning different fields of research. By allowing us to create and control entangled quantum states of light and matter, cavity quantum electrodynamics (QED) offers a fascinating platform in this context. This has led to several highly successful research directions, in fields as diverse as atomic physics [1–4], quantum information [5–8], and quantum fluids of polaritons [9–11]. Advances in controlling and probing light-matter interactions have allowed for the investigation of collective effects in solid-state systems such as atomically thin or layered materials [12–19]. Recently, pioneering work has also explored strong light-matter coupling in molecules and molecular solids [20–22].

One of the earliest and most important examples of collective phenomena in coupled light-matter systems is *superradiance*. Originally introduced by Dicke [23] in the description of the collective enhancement of spontaneous emission, superradiance signals a coherence in a ensemble of dipoles collectively interacting with the same radiation field. At equilibrium, superradiance appears as a phase transition [24,25] characterized by the macroscopic population of photons in the ground state and the collective ordering of dipoles induced by a photon-mediated effective dipole-dipole interaction. Analogous transitions have been studied in nonequilibrium conditions [26,27] and in the context of circuit QED [28–32].

In condensed matter physics, a wealth of emergent collective phases has been found in "quantum materials" [33], which result from strong interactions between electrons as well as other degrees of freedom. In this context, the engineering of new forms of effective interactions by means of collective light-matter coupling raises the fascinating possibility of exploring novel emergent collective phenomena involving entangled states of light and matter [34–38].

Littlewood and Zhu proposed early on [39] that superradiance may occur when electron-hole transitions in semiconducting quantum wells are coupled to a cavity photon. In their work, electronic interactions were not included and, most importantly, the photon diamagnetic term, which has been shown to impede superradiance [28,29,40], was neglected.

In this Letter, we demonstrate theoretically in a simple model of interacting electrons coupled to a cavity field how a cooperative effect between light-matter coupling and intrinsic electronic interactions can lead to the stabilization of a coherent light-matter phase, the "superradiant excitonic insulator" (SXI), characterized by the simultaneous appearance of equilibrium superradiance in the photon field and the condensation of excitons in the electronic system. As summarized in Fig. 1, superradiance cannot be reached in the absence of electronic interactions. At the same time coupling to the cavity promotes excitonic condensation in regimes in which it cannot be stabilized by electronic interactions only. Hence, SXI is a phase that intertwines superradiance and excitonic condensation in conditions where the two phases cannot be individually stabilized.

Light-matter Hamiltonian.—We consider spinless electrons moving in valence ($\nu = 1$) and conduction ($\nu = 2$) bands with hopping parameters $t_2 = -t_1 = t_{\mathrm{hop}}$, originating from localized atomic orbitals separated by an energy gap ω_{12}. The electrons interact via a local repulsive interaction U acting when two electrons sit on the same site. The electronic Hamiltonian reads:

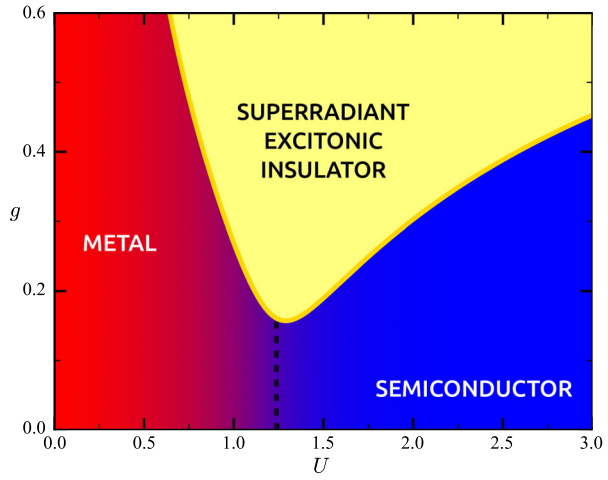

FIG. 1. Light-matter phases as a function of the strength of electronic interactions U and the light-matter coupling g at a temperature $T = 0.15$. The red (blue) intensity map reflects the evolution of the band populations. In the metal region (red) both orbitals are occupied, whereas in the semiconductor region (blue) the valence band is completely filled.

$$H_{el} = \sum_{\mathbf{k}\nu} \varepsilon_\nu(\mathbf{k}) c^\dagger_{\mathbf{k}\nu} c_{\mathbf{k}\nu} + U \sum_i n_{i1} n_{i2} - \mu \sum_{i\nu} n_{i\nu} \quad (1)$$

where $c^\dagger_{\mathbf{k}\nu}(c_{\mathbf{k}\nu})$ is the creation (annihilation) operator for an electron in the Bloch state $|\mathbf{k}\nu\rangle$ with quasimomentum $\mathbf{k}$ in the band ν and $n_{i\nu} = c^\dagger_{i\nu} c_{i\nu}$ is the electron number operator on lattice site i. While the results do not depend qualitatively on this choice, we consider for simplicity a one-dimensional lattice. The dispersion relations read $\varepsilon_2(\mathbf{k}) = -\varepsilon_1(\mathbf{k}) = (\omega_{12}/2) - 2t_{\mathrm{hop}} \cos k$ and we choose $\mu = (U/2)$ to fix the density to one electron per site $\langle n_{i1} + n_{i2} \rangle = 1$

The coupling of the electronic system [Eq. (1)] to an optical cavity is described by the light-matter Hamiltonian:

$$H = H_{el} + \omega_0 a^\dagger a + \Delta \hat{\rho} (a + a^\dagger)^2 + (a + a^\dagger) \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} \sum_{\nu\nu'} g_{\nu\nu'}(\mathbf{k}) c^\dagger_{\mathbf{k}\nu} c_{\mathbf{k}\nu'} \quad (2)$$

corresponding to a single-mode of frequency ω_0 with vector potential $\mathbf{A}(\mathbf{r}) \approx (\mathbf{A}_0/\sqrt{N})(a + a^\dagger)$ (dipole approximation) polarized along the spatial dimension of the electronic system. The third term in Eq. (2) is the diamagnetic contribution with $\Delta = [e^2/(2m)]\mathbf{A}_0^2$ and $\mathbf{A}_0 = \sqrt{[\rho/(2\omega_0 \epsilon_0)]}\mathbf{u}$, while the last term is the $\mathbf{k}$-dependent dipolar coupling between $|\mathbf{k}\nu\rangle$ Bloch states, $g_{\nu\nu'}(\mathbf{k}) = (e/m)\langle \mathbf{k}\nu | \mathbf{p} | \mathbf{k}\nu' \rangle \cdot \mathbf{A}_0 . \rho = N/V$ is the electronic density with V the cavity volume. ϵ_0 is the permittivity of the cavity and $\mathbf{u}$ the polarization vector.

The couplings of the diamagnetic and dipolar terms are not independent: they are related by the sum rules resulting from the canonical commutation relations $i = [\mathbf{r}, \mathbf{p}]$ [28,29]. In the Bloch basis we find that for interband transitions the following relation holds for each $\mathbf{k}$

$$\Delta = \sum_{\nu \neq \nu'} \frac{|g_{\nu\nu'}(\mathbf{k})|^2}{\varepsilon_{\nu'}(\mathbf{k}) - \varepsilon_\nu(\mathbf{k})}. \quad (3)$$

We assume to assign all the oscillators strength to the transition between the two low-energy bands $\nu = 1, 2$, obtaining

$$g_{12}(\mathbf{k}) = g f_{\mathbf{k}} \quad (4)$$

where $g \equiv \sqrt{\Delta \omega_{12}}$ is the light-matter coupling with dimension of an energy and $f^2_{\mathbf{k}} \equiv |\varepsilon_1(\mathbf{k}) - \varepsilon_2(\mathbf{k})|/\omega_{12}$ is a dimensionless factor characterizing the momentum dispersion. We neglect the intraband couplings as they do not play any role for the phase transition discussed here. The nature and validity of the approximations made are summarised in details in the Supplemental Material [41].

In the limit $U = t_{\mathrm{hop}} = 0$ the Hamiltonian [Eq. (2)] reduces to the well-known Dicke-Hopfield model of localized dipoles [23,45]. Dipole-dipole interactions between localized dipoles have been considered in Refs. [46–48]. For finite hopping, but $U = 0$, the model has been recently employed to describe charge transport in cavity-coupled semiconductors [49,50]. Energies are measured with respect to $\omega_{12} = 1$ and we fix $\omega_{12} = \omega_0$.

The Hamiltonian [Eq. (2)] has a global continuous symmetry $c_{i\nu} \to e^{i\varphi_\nu} c_{i\nu}$ and $a \to e^{i\lambda} a$ with $\varphi_1 - \varphi_2 = \lambda = \pm\pi$. We introduce two order parameters: the macroscopic expectation value of the photon field $\alpha \equiv \langle a \rangle/\sqrt{N}$ signaling superradiance [24,25], and $\Phi \equiv \langle c^\dagger_{i1} c_{i2} \rangle$ associated with the condensation of particle-hole pairs (excitons). The latter opens an hybridization gap, inducing an insulating phase known as an "excitonic insulator" (EI) [51–54]. A nonzero value of either α or Φ breaks the above symmetry.

In the thermodynamic limit for the electronic system, the light matter interaction can be treated exactly as the saddle point condition of an effective extensive action for the photon field

$$\Omega_0 \alpha + \frac{1}{N} \sum_{\mathbf{k}} \Gamma(\mathbf{k}) [\langle c^\dagger_{\mathbf{k}1} c_{\mathbf{k}2} \rangle_\alpha + \langle c^\dagger_{\mathbf{k}2} c_{\mathbf{k}1} \rangle_\alpha] = 0 \quad (5)$$

where $\Omega_0 \equiv \sqrt{\omega_0^2 + 4\Delta\omega_0}$ and $\Gamma(\mathbf{k}) \equiv g_{12}(\mathbf{k}) \sqrt{(\omega_0/\Omega_0)}$. The electronic averages $\langle \cdots \rangle_\alpha$ have to be computed with an effective interacting Hamiltonian $H^{\mathrm{eff}}_{el} = H_{el} + 2\alpha \sum_{\mathbf{k}} [\Gamma(\mathbf{k}) c^\dagger_{\mathbf{k}1} c_{\mathbf{k}2} + \mathrm{H.c.}]$ which is treated by introducing a Hartree-Fock (HF) decoupling of the interaction term $n_{i1} n_{i2} \to -m(n_{i1} - n_{i2}) - \Phi c^\dagger_{i2} c_{i1} - \Phi^* c^\dagger_{i1} c_{i2} + \mathrm{const}$, where $m \equiv \langle n_{i1} \rangle - \langle n_{i2} \rangle$ is the electronic orbital polarization.

Absence of superradiance.—The critical light-matter coupling g_c for superradiance is found by solving

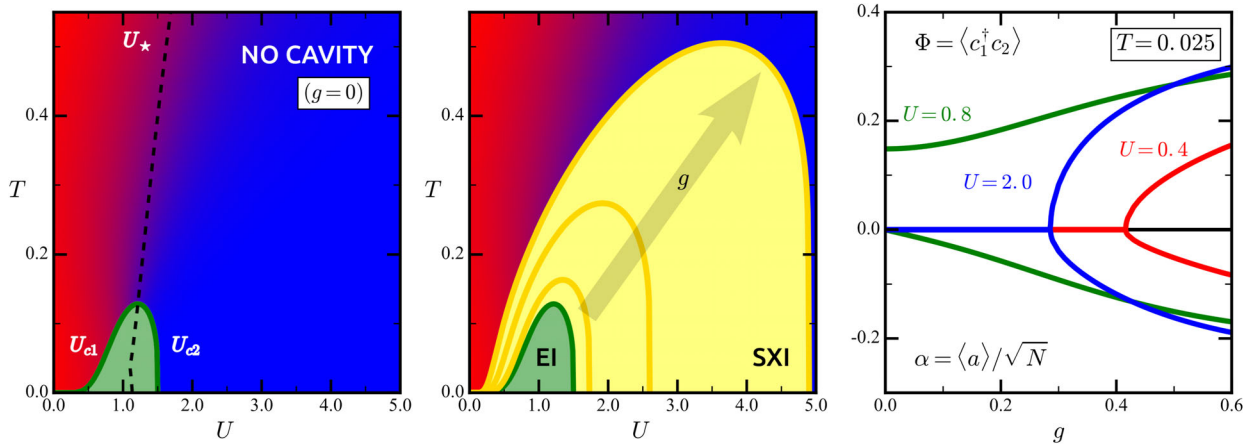

FIG. 2. (a) Phase diagram in the absence of light-matter coupling: metal (red), semiconductor (blue), and excitonic insulator (green). U_{c1} and U_{c2}: critical interactions for the EI. The red (blue) intensity map: orbital polarization as in Fig. 1. Dashed line: critical interaction for gap opening in the normal phase. (b) Phase diagram for increasing g (arrow): $g=0$ (green line) $g=0.2$, 0.4, and 0.6 (yellow lines). At finite g the EI (green) transforms into a SXI (yellow). (c) Excitonic and superradiant order parameters as a function of g at $T=0.025$ and three values of U.

Eq. (5) for g in the limit $\alpha \to 0^+$. In the case of noninteracting electrons $U=0$ at zero temperature this corresponds to the solution of

$$\Omega_0 - \frac{4}{N}\sum_{\mathbf{k}} \frac{\Gamma(\mathbf{k})^2}{|\varepsilon_1(\mathbf{k})-\varepsilon_2(\mathbf{k})|} = 0. \tag{6}$$

Using the definition of Ω_0 and $\Gamma(\mathbf{k})$, it is easy to see that this condition can never be satisfied for any value of g, electronic dispersions $\varepsilon_\nu(\mathbf{k})$ or $\omega_0 > 0$. This shows that no superradiant transition is possible: it is prevented by the diamagnetic coupling Δ that grows as g is increased [Eq. (4)]. This result extends to the case of itinerant electrons the "no-go theorem" for the Dicke transition [28,29,40]. The same result is obtained in the opposite atomic limit $t_{\rm hop} \to 0$ independently of electronic interactions [42,43].

Superradiant excitonic insulator.—The above picture dramatically changes once both electronic interactions U and electronic delocalization $t_{\rm hop}$ are taken into account. From now on we fix $t_{\rm hop}=0.5$. For $g=0$, Fig. 2(a), the electronic system is unstable towards the formation of an EI below a dome-shaped critical temperature $T_c(U)$ [51–54], between two critical interactions $U_{c1}(T) < U < U_{c2}(T)$ that merge at the maximum $T_{c,0}^{\rm max}$. At a high temperature, the system evolves from a metal ($0<m<1$) for $U<U_\star$ to a direct gap semiconductor ($m \simeq 1$) for $U>U_\star$ with a gap opening up between the two effective HF bands at $U=U_\star$.

At finite light-matter coupling, as expected by the above symmetry considerations, the EI phase transforms into a superradiant excitonic insulator (SXI), characterized by nonzero superradiant and excitonic order parameters [Fig. 4(c)]. This is easily understood as the EI ground state is characterized by a macroscopic electronic dipole moment [41,55]. Remarkably, when the system is not in the EI phase at $g=0$, there is (for $U \neq 0$) a critical value of the light-matter coupling g_c beyond which the SXI appears. As a result the phase space (temperature and interactions) for which the SXI is realized is significantly enhanced as the strength of the light-matter coupling is increased [Fig. 2(b)].

The occurrence of this intertwined light-matter collective phase is shown on Fig. 1, displaying the phase diagram as a function U and g for a temperature at which no coherent phase can be stabilized in the absence of light-matter coupling. The SXI phase intrudes between the metallic and semiconducting phases. At weak interaction $U<U_\star$, g_c increases upon decreasing U and diverges as $U \to 0$, as expected from the no-go theorem discussed above. On the contrary, in the strong interaction regime, g_c is an increasing function of U and approaches the finite value $g_c^\infty = \frac{1}{2}\sqrt{[(\omega_0\omega_{12})/(f_{\rm loc}^2-1)]}$ with $f_{\rm loc}=(1/N)\sum_{\mathbf{k}} f_{\mathbf{k}}$ as $U \to \infty$. At intermediate values of the interaction, corresponding to $U \sim U_\star$, the critical coupling has a minimum. By decreasing temperature such a dip in the phase boundary moves towards the $g=0$ axis until it becomes zero for $T=T_{c,0}^{\rm max}$ and splits into two points at $U=U_{c1}$ and $U=U_{c2}$, respectively, for $T<T_{c,0}^{\rm max}$.

Exciton-polariton softening.—The entangled nature of the SXI phase is investigated by looking at polariton modes resulting from the dressing of the cavity photon with the electronic transitions. This is characterized by the excitonic susceptibility $\chi(\omega)=\sum_{\mathbf{k}\mathbf{k}'}\chi^{12}_{\mathbf{k}\mathbf{k}'}(\omega)+\chi^{21}_{\mathbf{k}\mathbf{k}'}(\omega)$. Here, $\chi^{\nu\nu'}_{\mathbf{k}\mathbf{k}'}(\tau-\tau')=-\langle \mathcal{T}_\tau c^\dagger_{\mathbf{k}\nu}(\tau)c_{\mathbf{k}\nu'}(\tau)c^\dagger_{\mathbf{k}'\nu'}(\tau')c_{\mathbf{k}'\nu}(\tau')\rangle$ are the two-particles Green's functions computed in the random phase approximation (RPA) and in the absence of light-matter coupling:

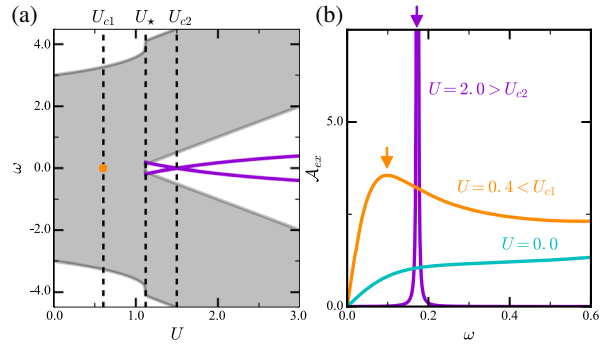

FIG. 3. (a) Spectrum of particle-holes excitations as a function of U at $T = 0.025$ at $g = 0$. The shaded area represents the particle-hole continuum. Dashed lines: U_{c1} and U_{c2}, and $U_\star$ as defined in Fig. 2(a). The dot at $U = U_{c1}$ indicates a $\omega = 0$ pole. Full lines: branches of poles crossing zero at $U = U_{c2}$. (b) Density of particle-hole excitations for three values of U. Arrows: excitonic resonances.

$$\chi_{\mathbf{k}\mathbf{k}'}(\omega) = \delta_{\mathbf{k}\mathbf{k}'}\chi^0_{\mathbf{k}}(\omega) - \frac{U}{N}\chi^0_{\mathbf{k}}(\omega)\sum_{\mathbf{q}}\chi_{\mathbf{q}\mathbf{k}'}(\omega) \qquad (7)$$

and $\chi^0_{\mathbf{k}}(\omega)$ the bare ($U = 0$) susceptibility.

The spectrum of particle-hole excitations as a function of the interaction U is displayed in Fig. 3. At $U = 0$ the spectrum is characterized by a featureless particle-hole continuum. At finite U we observe the formation of well-defined excitonic modes. At weak interaction $U < U_\star$ this mode corresponds to a resonance embedded in the particle-hole continuum, while for $U > U_\star$, an exciton gets split-off from the continuum and becomes a sharp pole inside the semiconducting gap [56].

At finite light-matter coupling, the particle-hole excitations hybridize with the photon through the polarization $\Pi(\omega) = (1/N)\sum_{\mathbf{k}\mathbf{k}'}\Gamma(\mathbf{k})\Gamma(\mathbf{k}')[\chi^{12}_{\mathbf{k}\mathbf{k}'}(\omega) + \chi^{21}_{\mathbf{k}\mathbf{k}'}(\omega)]$. This gives rise to the dressed photon spectra $\mathcal{A}_{ph}(\omega)$ displayed in Fig. 4 as a function of g for several values of U. In the noninteracting case $U = 0$ [panel (a)], the dressing produces two (lower and upper) polariton branches $\omega_\pm$ originating from the bare photon resonance. At finite interaction U, the lower branch shifts to low frequency, becoming a finite-width resonance for $U < U_\star$ [panel (b)] and a sharp mode for $U > U_\star$ [panel (c)]. Both modes become soft at the critical coupling g_c for the SXI transition.

At $U = 0$ the polariton modes result from the dressing by the continuum of bare particle-hole excitations. The latter are constrained by the sum-rules, which prevent the SXI transition and, in turn, the photon softening. On the contrary, at finite U, the lower polariton modes originate from the correlated excitonic modes discussed above [horizontal arrows in panels (b) and (c)]. Therefore, in the interacting case, the photon couples to an excitation of the many-body system and, as a result, the sum-rule for the bare particle-hole excitations no longer prevents a superradiant state which can indeed be reached at a finite critical value of the light-matter coupling.

The cooperation between the electronic interactions and light-matter coupling in the formation of the SXI state can be rationalized on the basis of a Landau expansion of the free-energy in terms of the two linearly coupled ordered parameters α and Φ, which reads: $F[\alpha, \Phi] = a_\alpha\alpha^2 + a_\Phi\Phi^2 + 2k\alpha\Phi + b\Phi^4 + \cdots$ with $a_\alpha > 0$ and $a_\Phi = c(T - T^0_c)$, with T^0_c the critical temperature associated with the uncoupled EI phase. At finite k, an instability related to a linear combination of the excitonic and superradiant eigenmodes occurs for $a_\alpha a_\Phi < k^2$, so that the SXI phase can be stabilized for $T > T^0_c$ for $k^2 > k^2_c = c(T - T^0_c)a_\alpha$ yielding an enhanced critical temperature $T_c = T^0_c + k^2/ca_\alpha$.

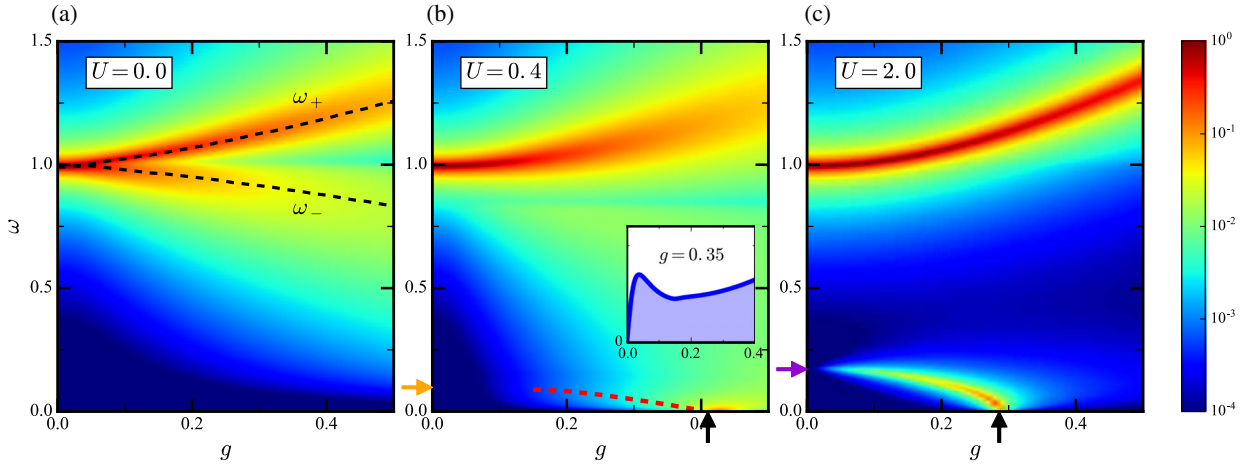

FIG. 4. Photon spectral functions $\mathcal{A}_{ph}$ as a function of the light-matter coupling g for $U = 0$ (a), $U = 0.4$ (b), and $U = 2.0$ (c) at $T = 0.025$. Dashed lines in panel (a): the upper and lower polaritons. Dashed line in panel (b): dispersion of the low-frequency resonance in the spectral function (see inset). Vertical arrows in panels (b) and (c) indicate the critical couplings for the SXI. Horizontal arrows: excitonic resonances at $g = 0$, as defined in Fig. 3(b).

The SXI transition does not rely on the pumping of photons into the cavity [57,58]. The coupling with an external environment, neglected here, is expected to bring only quantitative changes to the above physics such as shifting the critical couplings or temperatures [27,59]. However, this could play a crucial role in the observation of the SXI, in particular for the detection of the superradiant ground state [60] or the observation of the soft polariton modes [61].

In conclusion, we have investigated the cooperation between collective light-matter coupling and intrinsic electronic correlation. This leads to the SXI phase, entangling superradiance and excitonic condensation. Our results draw attention to quantum materials with strong electronic correlations as ideal test beds for the observation of entangled quantum states of light and matter. Specifically, collective light-matter coupling could be used as a probe for excitonic condensation in systems with a potential excitonic instability [62–64]. In this respect, we emphasize that recent experimental investigations of Ti_2NiSe_5 have suggested exciton condensation [64–68] in this material, and that optical excitations have revealed the interaction of the excitonic condensate with light [69–71]. Other possible candidates for exciton condensation providing a promising testing grounds for the SXI transition are electron-hole bilayers [72] such as bilayer graphene [73–77].

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