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 interactions in quantum materials and the long-range correlations 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.

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 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 \cite{1,4}, quantum information \cite{5,8} and quantum fluids of polaritons \cite{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 \cite{12,19}. Recently, pioneering work has also explored strong light-matter coupling in molecules and molecular solids \cite{20,22}.

One of the earliest and most important examples of the emergence of a collective ordered phase in cavity QED is the phenomenon of superradiance: the spontaneous emission of light occurring in an ensemble of two levels systems (dipoles) interacting with a photon field, as described by the Dicke model \cite{23-28}. The superradiant phase transition occurs due to the collective ordering of dipoles induced by a photon-mediated effective dipole-dipole interaction. Despite the fact that in most cases of physical interest such an ordered phase is not easily reached \cite{29-32}, this example shows how collective light-matter coupling can generate an effective interaction and quantum correlations in a many-body system.

In condensed matter physics, a wealth of emergent collective phases has been found in ‘quantum materials’ \cite{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 \cite{34-38}.

In this article, we demonstrate theoretically how this can lead to the stabilization of a coherent light-matter phase characterized by the simultaneous appearance of superradiance in the photon field and the condensation of excitons in the electronic system. We consider a simple model of an excitonic insulator, involving interactions between valence and conduction electrons \cite{39-42}. Additionally, the photon field of a cavity mediates a long-range light-matter dipolar coupling. Figure 1 provides an overview of our key finding: as a result of the cooperation between the correlations induced by the collective light-matter coupling and the electronic interactions intrinsic to the material, a ‘superradiant excitonic insulator’ (SXI) phase is stabilized. As shown below, superradiance cannot be reached in the absence of electronic interactions when only the coupling to the cavity field is present. At the same time, as clear from Fig. 1, the coupling to the cavity promotes excitonic condensation in regimes in which it cannot be stabilized by electronic interactions in the absence of the cavity. Hence, the light-matter coupling and electronic interactions display a cooperative effect which intertwines excitonic condensation and superradiance and leads to the SXI phase.

RESULTS

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

$$H_{el} = \sum_{k\nu} \varepsilon_{\nu}(k) c_{k\nu}^\dagger c_{k\nu} + U \sum_i n_i 1_n - \mu \sum_{i\nu} n_{i\nu} \quad (1)$$

where $c_{k\nu}^\dagger / c_{k\nu}$ is the creation/annihilation operator for an electron in the Bloch state $|k\nu\rangle$ with quasi-momentum $k$ in the band $\nu$ and $n_{i\nu} = c_{i\nu}^\dagger c_{i\nu}$ is the electron number operator on lattice site $i$. In the rest of the pa-
per we consider for simplicity a one-dimensional lattice. The presented results do not depend qualitatively on this choice. The dispersion relations read \( \epsilon_2(k) = -\epsilon_1(k) = \frac{\omega_2}{2} - 2t_{\text{hop}} \cos k \) and we choose the chemical potential \( \mu = \frac{U}{2} \) to fix the density to one electron per site \( (n_1 + n_2) = 1 \).

We imagine embedding the electronic system described by (1) inside an optical cavity. We consider the interaction with the electromagnetic field by assuming a single mode of frequency \( \omega_0 \) with vector potential polarized along the spatial dimension of the electronic system. By neglecting the spatial variation of the vector potential operator \( \mathbf{A}(r) \) (dipole approximation) the light-matter Hamiltonian is obtained after implementing the minimal substitution and expanding the electronic momentum operator \( \mathbf{p} \) in terms of Bloch states \( |k\nu\rangle \), yielding:

\[
H = H_{\text{EL}} + \omega_0 a^\dag a + \Delta (a + a^\dag)^2 + \frac{1}{\sqrt{N}} \sum_k (g(k) c_{k1}^\dag c_{k2} + h.c.).
\]

The first two terms are respectively the electrons and the photon Hamiltonians. The third term is the diamagnetic contribution with \( \Delta = \frac{e^2}{2m} A_0^2 \) and \( A_0 = \sqrt{\frac{e}{2\omega_0}} u \), while the last term is the \( k \)-dependent dipolar light-matter coupling with \( g(k) = \frac{e A_0}{m |k| |p|} k \cdot A_0 \). The electronic density is \( \rho = N/V \) with \( N \) the total number of electrons (equal to the number of lattice sites) and \( V \) the volume of the cavity. \( \epsilon_0 \) 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 \( i = [\mathbf{r}, \mathbf{p}] \) [31, 32]. When specialized to the two band case, this yields the relation (see Methods):

\[
g(k) = \gamma f_k \tag{3}
\]

where \( \gamma \equiv \sqrt{(\Delta \omega_{12})} \) is the light-matter coupling with dimension of an energy and \( f_k \) is a dimensionless factor that characterizes the momentum dispersion of the coupling. Its definition reads \( f_k \equiv |\epsilon_1(k) - \epsilon_2(k)|/\omega_{12} \) reducing to \( f_k = 1 \) in the case of localized electrons \( (t_{\text{hop}} = 0) \).

In the limit of fully localized and non-interacting electrons \( U = t_{\text{hop}} = 0 \) the Hamiltonian (2) reduces to the well known Dicke-Hopfield model [23, 43], with the (pseudo)spin 1/2 associated to the two possible states (valence or conduction) of the electron. For finite hopping, but in the absence of interactions \( (U = 0) \), this model has been recently employed to describe charge transport in cavity-coupled electronic systems [44, 45]. In the rest of the paper all the energies are measured with respect to the local splitting between the two bands \( \omega_{12} \) and we fix \( \omega_{12} = \omega_0 = 2t_{\text{hop}} \).

The Hamiltonian (2) has a global continuous symmetry involving the transformation of electronic and photonic operators according to \( c_{\nu} \rightarrow e^{i\varphi} c_{\nu} \) and \( a \rightarrow e^{i\lambda} a \) with \( \varphi_1 - \varphi_2 = \lambda = \pm \pi \). We introduce two order parameters: \( \alpha \equiv \langle a \rangle / \sqrt{N} \) associated with a macroscopic expectation value for the photon field signaling superradiance [24, 25], and \( \Phi \equiv \langle c_{1}^\dag c_{2} \rangle \) associated with the condensation of particle-hole pairs (excitons). The latter opens an hybridization gap between the valence and conduction bands, inducing an insulating phase known as an ‘excitonic insulator’ (EI) [39–42]. A non-zero value of either \( \alpha \) or \( \Phi \) breaks the above symmetry.

Taking the thermodynamic limit for the electronic system, the light matter interaction can be treated exactly: the partition function is dominated by the saddle point of the effective action for the photon field. The diamagnetic term \( (a^\dag + a)^2 \) can be eliminated by a canonical Bogoliubov transformation, leading to the rescaling \( \omega_0 \rightarrow \Omega_0 \equiv \sqrt{\omega_0^2 + 4\Delta \omega_0} \) and \( g(k) \rightarrow \Gamma(k) \equiv g(k) \sqrt{\frac{\omega_0}{\Omega_0}} \). We obtain the following self-consistent equation for \( \alpha \equiv \langle a \rangle / \sqrt{N} \):

\[
\Omega_0 \alpha + \frac{1}{N} \sum_k \Gamma(k) \left( \langle c_{k1}^\dag c_{k2} \rangle_\alpha + \langle c_{k2}^\dag c_{k1} \rangle_\alpha \right) = 0 \tag{4}
\]

where \( \langle \cdots \rangle_\alpha \) are averages computed with the effective
Hamiltonian

\[ H_{\text{el}}^{\text{eff}} = H_{\text{el}} + 2\alpha \sum_{k} (\Gamma(k) c_{k1}^\dagger c_{k2} + h.c.) \]  

(5)

The interacting electronic Hamiltonian (5) is treated by introducing a Hartree-Fock (HF) decoupling of the interaction term \( n_{k1} n_{k2} \rightarrow -m (n_{k1} - n_{k2}) - \Phi c_{k1}^\dagger c_{k2} - \Phi^* c_{k2}^\dagger c_{k1}^\dagger \) const where \( m \equiv (n_{k1}) - (n_{k2}) \) is the electronic orbital polarization. All three quantities \( \alpha, \Phi \) and \( m \) have to be determined self-consistently (see Methods).

Because \( \alpha \) and \( \Phi \) are linearly coupled, these two order parameters are expected to become unstable simultaneously, as observed in Ref. [46]. There, it was suggested that a superradiant transition may occur for non-interacting electrons and holes in parabolic bands coupled to a cavity field. In the following, we show that in fact this cannot happen in the absence of interactions, but that it becomes possible in the presence of both electronic interactions and light-matter coupling, for a wide range of parameters.

**Non-interacting electrons.** The critical light-matter coupling \( \gamma_c \) for superradiance is found by solving (4) for \( \gamma \) in the limit \( \alpha \rightarrow 0^+ \). In the case of non-interacting electrons \( U = 0 \) at zero temperature this corresponds to the solution of

\[ \Omega_0 - \frac{4}{N} \sum_{k} \frac{\Gamma(k)^2}{|\varepsilon_1(k) - \varepsilon_2(k)|} = 0. \]  

(6)

Note that, in this expression, both \( \Omega_0 \) and \( \Gamma(k) \) depend on the critical coupling \( \gamma_c \) through the definitions given above. Using those into equation (6) it is easy to see that this condition can never be satisfied for any value of \( \gamma_c \), electron dispersions \( \varepsilon_n(k) \) and \( \omega_0 \neq 0 \), showing the absence of any superradiant transition. In particular, superradiance is prevented by the diamagnetic coupling \( \Delta \) which, according to equation (3), becomes larger the larger the light-matter coupling \( \gamma \). We have thus extended to the case of itinerant electrons the so called ‘no-go theorem’ for the Dicke transition [29–32].

**Superradiant excitonic insulator.** The above picture dramatically changes once electronic interactions are taken into account. Fig. 2(a) displays the electronic phase diagram, in the absence of the cavity (\( \gamma = 0 \)). At \( U = 0 \) the system is a metal with two partially occupied overlapping bands \( m \leq 1 \). At high temperature, increasing \( U \) leads to a transition from the metal for \( U < U_4 \) to a direct gap semiconductor \( m \approx 1 \) for \( U > U_4 \), with a gap opening up between the two effective HF bands at \( U = U_4 \). Below a critical temperature \( T_e(U) \) the interaction \( U \) drives an instability towards the formation of the EI phase. The critical temperature is exponentially small at small \( U_4 \) goes through a maximum \( T_e^{\text{max}}(0) \) and vanishes as \( U \) is further increased. At a given temperature smaller than \( T_e^{\text{max}}(0) \) the EI phase exists for \( U_4(T) < U < U_2(T) \). These two critical interactions merge at the top of the dome for \( U \approx U_4 \). For \( U > U_2(0) \) no EI phase can be
stabilized at any temperature.

As the light-matter coupling is turned on, the EI becomes a superradiant excitonic insulator (SXI), characterized by non-zero superradiant and excitonic order parameters. The range of interaction strength and the temperature for which the SXI is realized are strongly enhanced as the light-matter coupling is increased (Fig. 2(b)). Panel (c) shows the evolution of the excitonic and superradiant order parameters as a function of the light-matter coupling at a fixed temperature, smaller than $T_{c,0}^{\text{max}}$. As expected, when the electronic system is already in the EI phase at $\gamma = 0$, the SXI is formed as soon as a non-zero $\gamma$ is turned on. Remarkably, when the system is not in the EI phase at $\gamma = 0$ there is (for $U \neq 0$) a critical value of the light-matter coupling $\gamma_c$ beyond which the SXI is realized.

Fig. 1 displays the phase diagram as a function $U$ and $\gamma$, for a temperature higher than $T_{c,0}^{\text{max}}$. It is seen that the light-matter coupling stabilizes the SXI phase, which in-\trudes between the metallic and semiconducting phases. At weak interaction $U < U_\ast$ the critical coupling $\gamma_c$ increases upon decreasing $U$, and diverges as $U \to 0$, as expected from the no-go theorem above. On the contrary, in the strong interaction regime, $\gamma_c$ is an increasing function of $U$ and approaches the finite value $\gamma_c^{\infty} = \frac{1}{2} \sqrt{\frac{2\Gamma_{\text{loc}}}{\hbar^2}}$ with $f_{\text{loc}} = \frac{1}{N} \sum_k j_k$ as $U \to \infty$. At intermediate values of the interaction, corresponding to $U \sim U_\ast$, the critical coupling has a minimum. By decreasing temperature such a dip in the phase boundary moves towards the $\gamma = 0$ axis until it becomes zero for $T = T_{c,0}^{\text{max}}$ and splits into two points at $U = U_{c,1}$ and $U = U_{c,2}$ respectively for $T < T_{c,0}^{\text{max}}$.

Exciton-polariton softening. Insight in the nature of the SXI transition can be reached by looking at the dressing of the cavity photon due to its coupling with the interacting electronic system.

The electronic interactions affect the photon dressing by modifying the spectrum of particle-hole excitations coupled to the light field. This is described by the excitonic susceptibility $\chi(\omega) = \sum_{kk'} \chi_{kk'}^{12}(\omega) + \chi_{kk'}^{11}(\omega)$. Here, $\chi_{kk'}^{\nu}(\tau - \tau') = -\langle T_{\tau} c_{kk'}(\tau)c_{kk'}(\tau')c_{kk'}(\tau)c_{kk'}(\tau') \rangle$ are the two-particles Green’s functions computed in the Random Phase Approximation (RPA):

$$\chi_{kk'}^{\nu}(\omega) = \delta_{kk'} \chi_k^0(\omega) - U \chi_k^0(\omega) \sum_q \chi_{qk'}(\omega)$$

and $\chi_k^0(\omega)$ the bare susceptibility. The spectrum of particle-hole excitations as a function of the interaction $U$ is displayed on 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 $U < U_\ast$ this mode corresponds to a resonance embedded in the particle-hole continuum, while for $U > U_\ast$, an exciton gets split-off from the continuum and becomes a sharp pole inside the semiconducting gap. In the absence of coupling with light, the excitonic instability occurring at $U = U_{c,1}$ is due to the above resonance becoming a zero-frequency pole, while that occurring at $U = U_{c,2}$ is driven by the softening of the exciton pole inside the gap (i.e. its frequency reaching zero). These two instabilities identify a BCS and a BEC regime of exciton condensation for $U < U_\ast$ and $U > U_\ast$ respectively.

At finite light-matter coupling, the excitonic modes hybridize with the photon, as illustrated on Fig. 4, which displays, for several values of $U$ and $\gamma$, the dressed photon spectral function $A_p(\omega) = -\frac{1}{2} \text{Im} D_{aa}(\omega)$, with $D_{aa}$ the photon Green’s function (see Methods). In the non-interacting case $U = 0$ (panel (a)), the dressing by the continuum of particle-hole excitations produces two (lower and upper) polariton branches. This is similar to Dicke-like models and we note that these polariton peaks are centered at finite frequencies $\omega_{\pm}$ for any value of the light-matter coupling $U_{c,0}$. At finite interaction $U$, the lower branch shifts to low frequency, becoming a finite-width resonance for $U < U_\ast$ (panels (b) and (e)) and a sharp mode for $U > U_\ast$ (panels (c) and (f)). By following their dispersions as a function of $\gamma$, we identify these excitations as exciton-polaritons originating from the interaction-induced excitonic modes discussed above (horizontal arrows in panels (b) and (e)). The key point, illustrated by the vertical arrows in panels (b) and (e) of
FIG. 4. Exciton-Polariton Softening. Top panels: Photon spectral functions $A_{ph}$ as a function of the light-matter coupling $\gamma$ for $U = 0$ (a), $U = 0.4$ (b) and $U = 2.0$ (c). The intensities of the spectral functions have been normalized with respect to $A_{ph}^{\text{max}}$ corresponding to the maximum value of the intensity in the absence of light-matter coupling. Vertical arrows in panels (b) and (c) indicate the critical couplings for the SXI. These corresponds to a zero frequency resonance for the dressed photons. Bottom panels: Photon spectral functions for the corresponding values of $U$ in the top row at the fixed values of light-matter coupling indicated in each panel. The inset in panel (e) is a blow-up of the finite-width exciton-polariton resonance at low frequency. The dispersion of this mode as a function of $\gamma$ is displayed by the red dashed line in panel (b). Horizontal arrows in panels (b)-(c) indicate the energy of the excitonic resonance at $U = 0.4$ and of the excitonic bound state at $U = 2.0$.

Fig. 4 is that, in both regimes, the SXI transition corresponds the softening of these exciton-polaritons modes at the related critical value of the light-matter coupling.

DISCUSSION

Our results highlight the crucial role of interactions between electrons in stabilizing the coherent light-matter state discussed in this article. Indeed, in the absence of interactions, light couples to the bare electron-hole excitations, whose matrix elements are constrained by the sum-rule discussed above. As a result, a superradiant phase cannot be realized. In contrast, in the presence of interactions, light couples to the excitonic mode, which is an excitation of the many-body system. While the sum-rule still holds for dipolar transitions between the bare states, it no longer prevents a superradiant state which can indeed be reached at a finite critical value of the light-matter coupling. At this critical point, the dressed photon excitation becomes soft.

We notice that the exciton-polariton softening does not require the tuning of the photon frequency to the energy of the excitonic mode. In fact, throughout the paper we fixed the photon frequency $\omega_0$ to the average energy of the bare electronic transitions, $\omega_{12}$, which can be largely detuned from the interaction dependent energy of the excitonic mode. We observe that, despite such a large detuning, the light strongly hybridizes with the exciton mode, eventually leading to the soft polariton at the SXI transition.

The fact that the SXI state results from a cooperation between electronic interactions and the light-matter coupling can be rationalized on the basis of a Landau
expansion of the free-energy in terms of the two linearly coupled ordered parameters $\alpha$ and $\Phi$, which reads:

$$F[\alpha, \Phi] = a_\alpha \alpha^2 + a_\Phi \Phi^2 + 2k_0 \alpha \Phi + b \Phi^4 + \cdots.$$  

In this expression $a_\alpha$ is positive, reflecting the fact that no superradiance can occur in the absence of coupling to the excitonic order parameter (i.e. when $k = 0$). In contrast, $a_\Phi = c(T - T_c^0)$, with $T_c^0$ the critical temperature associated with the EI phase in the absence of the cavity. Analyzing this expression, it is easily seen that in the presence of the coupling $k$ between the two order parameters, an instability occurs for $a_\alpha a_\Phi < k^2$. Hence, the critical temperature is enhanced by the coupling to the cavity, according to: $T_c = T_c^0 + k^2 / a_\alpha$. Equivalently, the SXI phase can be stabilized for $T > T_c^0$ for $k^2 > k^2_c = c(T - T_c^0) a_\alpha$. This highlights the fact that the regime of stability of the SXI phase is enhanced by the light-matter coupling as compared to that of the EI phase, as found above. Furthermore, the unstable mode is a linear combination of the excitonic and superradiant eigenmodes, and hence has both $\alpha \neq 0$ and $\Phi \neq 0$. Identical arguments hold in the case light-matter coupling drives a zero temperature quantum phase transition for $U > U_c(0)$.

The cooperative effect between collective light-matter coupling and intrinsic electronic interactions draws the attention to quantum materials with strong electronic correlations as ideal test beds for the observation of entangled quantum states of light and matter. In this context, layered van der Waals materials have recently attracted considerable attention due to their low dimensionality and their peculiar optical properties. These materials have been incorporated in photonic and plasmonic devices and strong light-matter coupling has been observed [12, 13]. These materials also exhibit strong many body effects, highlighted for example by large excitonic binding energies in layered transition-metal dichalcogenides [14–19].

The SXI transition predicted in this paper suggests the exploration of collective light-matter coupling in systems with a potential excitonic instability [48, 49]. We emphasize in particular that recent experimental investigations of Ti$_2$NiSe$_5$ have suggested exciton condensation [49–51] in this material, and that nonlinear laser excitations have revealed the interaction of the excitonic condensate with light [52–54]. Other possible candidates for exciton condensation which may provide promising testing grounds for the SXI transition are electron-hole bilayers [55] such as bi-layer graphene [56–58]. Recent experimental evidence of effects associated with strong electronic correlations in bi-layer graphene is promising in this respect [59, 60].

In summary, we have investigated the cooperative effects between collective light-matter coupling and intrinsic electronic correlations in interacting electron systems coupled to a cavity photon. We have shown that this leads to a light-matter coherent phase, the superradiant excitonic insulator (SXI), which entangles superradiance and exciton condensation. The SXI phase can be stabilized over a much larger range of temperatures and interaction strengths than the excitonic insulating phase in the absence of light-matter coupling. The electronic interactions stabilize superradiance, while it is prevented by diamagnetic coupling in the absence of interactions. These results open intriguing perspectives for the investigation of effects associated with strong light-matter coupling in quantum materials.

**METHODS**

**Light-Matter Hamiltonian and Sum Rules for Bloch-Electrons** Spinless electrons in a periodic potential provided by a crystalline lattice $U(r)$ move accordingly to the non-interacting Hamiltonian

$$H_0 = \sum_j \frac{p_j^2}{2m} + U(\{r\}).$$  

(8)

The light-matter interaction is described by the minimal substitution $p_j \rightarrow p_j + eA(r_j)$ where $p_j$ is the momentum of the $j-th$ electron and

$$A(r) = \sqrt{\frac{1}{2\omega_0\epsilon_0}} V (e^{iqr} a + e^{-iqr} a^\dagger).$$  

(9)

the vector potential for the single mode in the cavity. In the dipole approximation $A(r) \approx \frac{\mathbf{A}_0}{\sqrt{N}} (a + a^\dagger)$, with $\mathbf{A}_0$ defined in the main text, we get

$$H_{lm} = H_0 + \frac{1}{\sqrt{N}} \sum_j \frac{e}{m} p_j \cdot \mathbf{A}_0 (a + a^\dagger) + \frac{e^2}{2m} \mathbf{A}_0^2 (a + a^\dagger)^2 + \omega_0 a^\dagger a.$$  

(10)

The electronic part of the Hamiltonian is expanded on the basis of Bloch states which diagonalize the bare Hamiltonian $H_0 |kv\rangle = \varepsilon_{kv}(k) |kv\rangle$. By introducing creation/annihilation operators $c_{kv}^\dagger/c_{kv}$ for electrons with quasi-momentum $k$ in the $\nu$ band we get

$$H_{lm} = \sum_{kv}\varepsilon_{kv}(k) c_{kv}^\dagger c_{kv} + \frac{(a + a^\dagger)}{\sqrt{N}} \sum_{k} \sum_{\nu\nu'} g_{\nu\nu'}(k) c_{kv}^\dagger c_{kv'} + \frac{e^2}{2m} \mathbf{A}_0^2 (a + a^\dagger)^2 + \omega_0 a^\dagger a.$$  

(11)

where we have defined the $k-$dependent dipole matrix element as $g_{\nu\nu'}(k) = \frac{e}{\sqrt{N}} p_{\nu\nu'}(k) \cdot \mathbf{A}_0$ with $p_{\nu\nu'}(k) = \langle kv|p|kv'\rangle$. In deriving the expression for $g_{\nu\nu'}(k)$ we expanded Bloch states in terms of Wannier wave functions $w_{R\nu}(r)$ and used the translational properties $w_{R\nu}(r) = w_{R+\mathbf{R}\nu}(r + \mathbf{R})$. 


Following Ref. [31] we relate the matrix elements \( g_{\nu \nu'}(k) \) to the diamagnetic coupling \( \Delta = \frac{e^2}{2m} A^2 \). By using sum rules deriving from the fundamental commutation relation \([r, p] = i\), we obtain that for fixed value \( \Delta \) and for each \( k \) and \( \nu \) the couplings \( g_{\nu \nu'}(k) \) fulfill the relation

\[
\Delta = \sum_{\nu \neq \nu'} \frac{|g_{\nu \nu'}(k)|^2}{\varepsilon_{\nu'}(k) - \varepsilon_{\nu}(k)}. \tag{12}
\]

In the present case we consider that the light field couples only with two low-energy bands which are well separated by the rest of all the high-energy bands. In such a situation the sum rule (12) is almost saturated by restricting to \( \nu = 1, 2 \). By applying the sum rule for each \( k \) to the band with the lowest energy we get the coupling in equation 3.

**Saddle point equation and Hartree-Fock approximation** The saddle point equation is derived by formally integrating the fermionic fields in an imaginary time path integral formalism. This leads to the effective extensive action for the photon fields

\[
S_{\text{eff}}[\phi^*, \phi] = N\int d\tau d\tau' \phi^*(\tau) D_0^{-1}(\tau - \tau') \phi(\tau) - \log Z_0(\phi, \phi^*) = N\sigma_{\text{eff}}[\phi^*, \phi], \tag{13}
\]

where \( Z_0(\phi, \phi^*) \) is the fermionic partition function depending on the photonic fields \( \phi(\tau) \) and \( D_0^{-1} \) the inverse of the bare photon propagator. In the large \( N \) limit the partition function is dominated by the saddle-point of the effective action. By taking the variation \( \delta \sigma_{\text{eff}} = 0 \) assuming time independent fields \([61]\), \( \phi(\tau) = \alpha \), we obtain the saddle point equation (4).

The saddle point equation (4) depends on the fermionic expectation values \( \langle c_{k,1}^\dagger, c_{k,2} \rangle_\alpha \) for an effective interacting electronic model determined by the value of the superradiant order parameter \( \alpha \) (5). The electronic problem is treated in the Hartree-Fock approximation by decoupling the interacting part of the Hamiltonian

\[
n_1 n_2 \rightarrow \langle n_{11} \rangle n_{12} + \langle n_{12} \rangle n_{11} - \langle c_{i1}^\dagger c_{i2}^\dagger \rangle c_{i2}^\dagger c_{i1} - \langle c_{i2}^\dagger c_{i1} \rangle c_{i1}^\dagger c_{i2} \tag{14}
\]

in terms of the mean-field orbital polarization \( m \) and excitonic amplitude \( \Phi \)

\[
m \equiv \langle n_{12} \rangle - \langle n_{11} \rangle \tag{15}
\]
\[
\Phi \equiv \langle c_{i1}^\dagger c_{i2} \rangle
\]

The Hartree-Fock Hamiltonian is easily diagonalized and the values \( m \) and \( \Phi \) are obtained by imposing the self-consistency relations (15). Explicit equations read

\[
m = \frac{1}{N} \sum_k \delta f(k) \frac{\omega_-(k)}{E(k)} \tag{16}
\]
\[
\Phi = \frac{1}{N} \sum_k \delta f(k) \frac{\lambda(k)}{2E(k)}
\]

where \( \delta f(k) = f[\omega_-(k) + E(k)] - f[\omega_+(k) - E(k)] \), with \( f(x) \) the Fermi distribution function at temperature \( T \), \( E(k) = \sqrt{\omega_-(k)^2 + \lambda(k)^2} \), with \( \omega_+(k) = \frac{\varepsilon_2(k) + \varepsilon_1(k)}{2} \) \( \omega_-(k) = \frac{\varepsilon_1(k) - \varepsilon_2(k)}{2} - \frac{i}{2} m \) and \( \lambda(k) = 2\Gamma(k) \alpha - U\Phi \).

**Gaussian fluctuations and photon dressing** The photon dressing discussed in Fig. 4 is obtained by taking fluctuations around the saddle point condition corresponding to the normal phase (i.e. \( \alpha = 0 \)) \([62, 63]\). This gives a Dyson equation for the photon propagator \( D(\omega) = D_0(\omega) + D_0(\omega)\Pi(\omega)D(\omega) \) where \( D_0(\omega) \) and \( D(\omega) \) are the free and dressed photon propagator in Nambu notation \( D(\omega) = (D_{aa}(\omega) D_{ab}(\omega)) \) and \( D_{ab}(\omega) = -i \int d\tau e^{i\omega\tau} \delta(t) \langle [A(t), B] \rangle \).

The polarization \( \Pi(\omega) \) that characterizes the photon dressing is the same for all four components and reads

\[
\Pi(\omega) = \frac{1}{N} \sum_{kk'} \Gamma(k) \Gamma(k') \chi_{kk'}(\omega), \tag{17}
\]

with \( \chi_{kk'}(\omega) \) the excitonic susceptibilities at zero light-matter coupling computed in the Random Phase Approximation, as defined in the main text.

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