Mediated interactions and photon bound states in an exciton-polariton mixture

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The quest to realise strongly interacting photons remains an outstanding challenge both for fundamental science and for applications. Here, we explore mediated photon-photon interactions in a highly imbalanced two-component mixture of exciton-polaritons in a semiconductor microcavity. Using a theory that takes into account non-perturbative correlations between the excitons as well as strong light-matter coupling, we demonstrate the high tunability of an effective interaction between quasiparticles formed by minority component polaritons interacting with a Bose-Einstein condensate (BEC) of a majority component polaritons. In particular, the interaction, which is mediated by the exchange of sound modes in the BEC can be made strong enough to support a bound state of two quasiparticles. Since these quasiparticles consist partly of photons, this in turn corresponds to a dimer state of photons propagating through the BEC. This gives rise to a new light transmission line where the bound state wave function is directly mapped onto correlations between outgoing photons. Our findings open up new routes for realising highly non-linear optical materials and novel hybrid light-matter quantum systems.

The goal to achieve strong photon-photon interactions motivates the intense investigation of novel quantum materials and drives forward our fundamental understanding of quantum states of light. Exciton-polaritons, in short polaritons, are hybridised states of light and excitons in semiconductors inside microcavities that have risen as a promising platform to realise such strong interactions thereby providing a pathway to highly non-linear optics with a range of technological applications [1–13]. In spite of impressive experimental progress [14, 15], it has, however, turned out to be difficult to make the photon-photon interaction sufficiently strong to realise these objectives. Mechanisms to increase the interaction strength include Feshbach resonances [16–19], dipolar excitons [20, 21], strongly correlated electrons [22], and excitons in Rydberg states [23].

Recently, it has observed the formation of quasiparticles, coined polaron-polaritons, resulting from the interaction between the excitonic part of the polariton and a surrounding medium consisting either of excitons in another spin state [16, 19] or electrons [24, 25]. An inherent feature of quasiparticles is that they interact via the exchange of density modulations in the surrounding medium. Such mediated interactions give rise to a range of important many-body phenomena establishing e.g. the realm of Landau’s liquid theory [26, 27], leading to conventional [28] and high Tc superconductivity [29], and the fundamental interaction in particle physics [30].

Here, we explore mediated interactions in a highly imbalanced two-component mixture of polaritons created by a pump-probe scheme inside a two-dimensional (2D) semiconductor microcavity as illustrated in Fig. 1(a). We develop a strong coupling theory describing the effective interaction between two quasiparticles formed by minority component polaritons interacting with a surrounding BEC of the majority polaritons. This interaction is shown to be attractive and tuneable and as a striking consequence, it supports bound states of two quasiparticles. Since these dimer states partly consist of two photons, their propagation through the BEC leads to the emergence of an additional line in the light transmission spectrum, see Fig. 1(a). The dimer wave function is moreover shown to be imprinted on the correlations of the transmission.

FIG. 1: (a) A pump beam creates a BEC of exciton-polaritons (red balls) inside a 2D semiconductor in a microcavity. A probe beam with different polarisation creates quasiparticles called polaron-polaritons (gray balls), which can bind via an effective interaction mediated by the BEC to form dimer states. This gives rise to a distinct line of light transmission carried by the dimer states involving two photons propagating through the BEC. The red, green, and blue lines show energy of the polaritons, the polaron-polaritons, and the dimers respectively as a function of the detuning δ. (b) The Landau effective interaction between the polaron-polaritons.
mitted photons allowing for a direct detection.

System.– We consider a 2D mixture of exciton-polaritons in spin states $\sigma = \uparrow, \downarrow$. The Hamiltonian is

$$\hat{H} = \sum_{k,\sigma} \left[ \varepsilon_{\uparrow k} \hat{c}_{\uparrow k}^\dagger \hat{c}_{\uparrow k} + \varepsilon_{\downarrow k} \hat{c}_{\downarrow k}^\dagger \hat{c}_{\downarrow k} \right] + g_\uparrow \sum_q \hat{\rho}_{\uparrow q} \hat{\rho}_{\uparrow q} + \frac{g_\downarrow}{2} \sum_q \hat{\rho}_{\downarrow q} \hat{\rho}_{\downarrow q},$$  \tag{1}

where $\hat{c}_{\sigma k}^\dagger$ creates an exciton with 2D transverse momentum $k$, spin $\sigma$, and kinetic energy $\varepsilon_{\sigma k} = k^2/2m_\sigma$. Likewise, $\hat{c}_{\sigma k}$ creates a photon with momentum $k$, spin $\sigma$, and kinetic energy $\varepsilon_{\sigma k} = k^2/2m_\sigma + \delta$, where $\delta$ is the detuning. We have defined $\hat{\rho}_{\sigma q} = \sum_k \hat{\rho}_{\sigma k-q} \hat{\rho}_{\sigma k}$ and used units where the system volume and $\hbar$ are one. The first line of Eq. (1) describes excitons coupled to photons in the microcavity with Rabi frequency $\Omega$, giving rise to the formation of lower and upper exciton-polariton branches with energies $\varepsilon_{\downarrow k} = \varepsilon_{\uparrow k} + \delta_k = \sqrt{\varepsilon_{\uparrow k}^2 + 4\Omega^2}/2$, where $\delta_k = \varepsilon_{\downarrow k} - \varepsilon_{\uparrow k}$.

The second line in Eq. (1) describes the interaction between excitons with opposite and parallel spins with strengths $g_\uparrow$ and $g_\downarrow$ respectively. They are both taken to be momentum independent, since their typical length scale is given by the exciton radius, which is much shorter than any other relevant length scale. Whereas the interaction between parallel spins is weak, we consider the case where the interaction between excitons with opposite spins leads to the formation of a bi-exciton state as observed experimentally [16, 19, 31, 32]. This gives rise to a Feshbach resonance, which is described by the scattering matrix $\mathcal{T}(p)$, where $p = (p, \omega)$ is the total momentum/energy of the $\uparrow$ and $\downarrow$ scattering pair [33].

As illustrated in Fig. 1(a), we consider a BEC of exciton-polaritons with density $n_B$ and spin-polarization $\uparrow$ created by a pump beam. A weaker probe beam creates a small density of exciton-polaritons with spin-polarization $\downarrow$, which can be regarded as impurities. Their interaction with the surrounding BEC leads to the formation of quasiparticles coined polaron-polaritons [34, 35], due to their strong similarities with polaron formation in atomic gases [36–38]. In Fig. 1(a), the green line shows the energy of the lowest polaron-polariton branch as a function of the detuning $\delta$. It is lowered compared to the lower polariton energy $\varepsilon_{\uparrow k} = (\varepsilon_{\uparrow k} + \varepsilon_{\uparrow k} - \sqrt{\varepsilon_{\uparrow k}^2 + 4\Omega^2})/2$ due to interactions with the BEC of $\uparrow$ polaritons. The energy of the polaron-polariton branch in Fig. 1(a) was calculated using a ladder approximation generalised to include the strong light-matter coupling. This gives rise to a matrix structure of the photon and exciton Green’s functions coupled by light with a self-energy in the exciton channel of the form [33, 34]

$$\Sigma_{\sigma}(p) = n_B C_q^2 \mathcal{T}(p),$$  \tag{2}

where $C_q^2 = (1 + \delta_q/\sqrt{\delta_q^2 + \Omega^2})/2$ is the Hopfield coefficient of the polaritons in the BEC [34, 39]. In the calculation, we used realistic experimental parameters with a Rabi splitting $\Omega = 3.5$ meV, exciton mass $m_\pi = 0.16m_e$ with $m_e$ the electron mass, and $m_\pi = 10^{-4}m_e$ [15, 16, 40–42]. The density of the BEC is $n_B = 3.5 \times 10^{15}$ cm$^{-2}$, the direct exciton-exciton coupling $g_{\uparrow \uparrow} \approx 3 \mu$eV$\mu$m$^{-2}$, and the energy of the bi-exciton state is $\varepsilon_{\uparrow \uparrow} = 0.8$ meV.

Effective interaction.– Our focus here is on the interaction between polaron-polaritons mediated by sound modes in the BEC. We first calculate the mediated interaction between two $\downarrow$ bare excitons using a non-perturbative approach that includes strong Feshbach correlations between a pair of $\uparrow \downarrow$ excitons exactly, combined with Bogoliubov theory generalised to the steady-state BEC at hand [2, 43, 44]. The mediated interaction between two $\downarrow$ excitons with energy/momentum $p - q/2$ and $p' + q/2$ scattering into final states with energy/momentum $p + q/2$ and $p' - q/2$ is

$$V(p, p'; q) = n_B C_q^2 C_q^2 \mathcal{T}^*(p) G^{LP}(q) \mathcal{T}(p').$$  \tag{3}

Here $G^{LP}(q)$ is a $2 \times 2$ matrix containing the normal and anomalous Green’s functions describing sound propagation in the BEC [33]. We have defined the vector $\mathcal{T}^*(p) = [\mathcal{T}(p + q/2) \mathcal{T}(p - q/2)]$ describing the scattering between the sound mode and the excitons, and the Hopfield factors in Eq. (3) are due to the fact that it is only the excitonic part of the BEC that scatters. The factor $n_B$ reflects that the interaction is mediated by the BEC. Details on the derivation of Eq. (3) are given in the Supp. Mat. [33]. The mediated interaction $V(p, p'; q)$ depends on both the incoming as well as the transferred momenta due to the lack of Galilean invariance. It also depends on their energies due to the finite speed of sound in the BEC giving rise to retardation effects.

Having obtained the mediated interaction between two $\downarrow$ excitons, we can now calculate the effective interaction between the polaron-polariton quasiparticles, which is the physically relevant quantity. As in the derivation of Landau’s Fermi liquid theory [27, 45], this is obtained by evaluating the mediated interaction on-shell between two polaron-polaritons taking into account their quasiparticle residues in the exciton channel. Consider for concreteness the scattering between two polaron-polaritons in the branch shown by the green line in Fig. 1(a). Taking both the incoming and outgoing momenta to be zero, we obtain from Eq. (3)

$$f(0,0) = \frac{2T^2(0,\varepsilon_0)Z_0^4}{3g_{\uparrow \uparrow}},$$  \tag{4}

where $\varepsilon_0$ and $Z_p$ denote the energy and exciton residue of a polaron-polariton with momentum $p$. The effective interaction in Eq. (4) depends on the $\downarrow \uparrow$ scattering matrix $\mathcal{T}$ squared, reflecting the basic mechanism is the emission and subsequent absorption of a sound mode in the BEC. Also, the $1/g_{\uparrow \uparrow}$ dependence shows that the interaction is stronger the more compressible the BEC. Compared to the mediated interaction between two impurities interacting with a conventional BEC [46–48], Eq. (4) contains an additional factor 2/3 originating from the non-equilibrium nature of the BEC, as well as the residue $Z_p$ giving the exciton component of the polaron-polariton.
Assuming a polaron-polariton density \( \delta \) shift \( n_f \) which in the ladder approximation reads \([49]\)

\[ n_f \text{ bi-exciton with increasing attractive lower polaron-polariton approaches that of the momentum/energy dependence of the mediated interaction between a pair of ↓ excitons. A systematic capture such dimers, we must describe strong correlations of bound states consisting of two polaron-polaritons. To geometry, this naturally raises the intriguing possibility prolonged by the coherence length of the BEC and the 2D case is due to many-body effects, which alter the one particle dispersion into that of a polaron-polariton and make the mediated interaction non-local, i.e. depending on all momenta \([51]\).}

**Light transmission.** – We now show explicitly how the formation of dimer states gives rise to a distinct line of transmitted light. To do so, the strong correlations due dimer formation via the mediated interaction must be included when calculating the photon propagator. In Landau theory, the energy shift of a quasiparticle with momentum \( p \) due to an effective interaction \( f(p, k) \) with other quasiparticles with population \( \delta n_k \) is given by \( \delta \varepsilon_p = \int f(p, k) \delta n_k d^3 k/4\pi^2 \) \([27]\). Following this, the self-energy due to the mediated interaction in the exciton channel has the form

\[ \Sigma_{in}(p) = nZ_p [\Gamma(p, 0; 0) + \Gamma(p, 0; -p)], \]

where we have assumed a density \( n \) of polaron-polaritons with zero momentum. The two terms in Eq. \((6)\) are the energy shifts coming from the mediated interaction in the Hartree and Fock channels generalised to include strong coupling effects using the BSE \([33, 49]\), as shown in Fig. 2.

![FIG. 2: The ↓ exciton self-energy with the first term describing scattering with the BEC giving Eq. (2). The other terms correspond to Eq. (6) giving scattering between two ↓ excitons via the mediated interaction leading to dimer formation. Red lines are the ↓ exciton propagator, dashed lines are ↑ polaritons in the BEC, the box is the \( T \) matrix, and the wavy line is the mediated interaction in Eq. (3). The first order term in the mediated interaction is included in the \( T \) matrix.](image)

Adding \( \Sigma_{in}(p) \) to Eq. \((2)\) includes dimer formation in our many-body theory. A pole in \( \Gamma \) coming from the presence of a dimer will show up as a pole in both the exciton and photon propagators as they are coupled by the Rabi matrix element \( \Omega \). Further details of our theory are given in the Supp. Mat. \([33]\).}

In Fig. \(3\), we plot the ↓ photon spectral function as a function of the detuning \( \delta \) calculated numerically for the same parameters employed in Fig. \(1\). There are several polaron-polariton branches typical of the interplay between strong interactions and light coupling \([24, 25, 34, 35]\). The lowest quasiparticle branch, which we assume
to be populated with density \( n = 0.15 n_E \) corresponds to the green line in Fig. 1(a). There is however one distinct line in the spectrum, marked as ”dimer”, which is not there for zero \( n = 0 \). This transmission line comes from the fact that when the energy of the incoming photon and a polaron-polariton already present equals that of a dimer state, a bound state involving two photons is formed, which propagates through the BEC giving rise to light transmission. The dimer state also gives rise to an avoided crossing feature in the spectrum, which becomes more pronounced the larger the polaron-polariton density \( n \). In an experiment, one could for instance use a spectrally broad probe beam creating both the dimer states and the polaron-polaritons from which they are formed. The intensity of the dimer transmission line will scale as the intensity of the probe beam squared due to the two-body nature of dimer formation. Note that the effective interaction between the other polaron-polariton branches in Fig. 3 is much weaker than that for the lowest branch since they are further away from the Feshbach state, and there are therefore no other dimer lines.

**Photon correlations.**—We finally show that the wave function of the dimer is imprinted in the transmitted light correlations. Figure 4 plots the correlation function \( g_2(p, -p) = \langle a_p a_p a_p^\dagger a_p^\dagger \rangle - \langle a_p a_p \rangle \langle a_p^\dagger a_p^\dagger \rangle \) for different detunings \( \delta \), where, \( a_p^\dagger \) creates a polaron-polariton with momentum \( p \). It is calculated using the wave function \( |\Phi\rangle = \sum_{p>0} \phi(p) a_p^\dagger a_p^\dagger |0\rangle \) of the dimer as \( g_2(p, -p) = |\phi(p)|^2 - |\phi(p)|^4 \), where \( |0\rangle \) is the vacuum state. Here, \( \phi(p) \) is obtained from the BSE by mapping it onto an effective Schrödinger equation [33, 51]. We see that \( g_2 \) indeed is non-zero when there is a bound state. The correlations increase with increasing \( \delta \) as the dimer becomes more deeply bound. The kink around \( p \approx 0.05 k_n \) reflects that above this momentum, the polaron-polaritons become almost pure excitons with a much larger mass. Importantly, Fig. 4 demonstrates that the wave function of the bound state can be measured directly from the correlations of the transmitted light.

**Discussion.**—Since we are considering a strongly correlated hybrid light-matter system, it is worth to discuss the accuracy of our approach. First, the ladder approximation describing the formation of polaron-polaritons is surprisingly accurate for the analogous problem of atomic polaron formation [36, 37, 52–55]. An analogous theory for the mediated interaction between impurities and dimer formation in an atomic BEC has been shown to be remarkably accurate even for strong interactions when benchmarked against Monte-Carlo calculations [48, 51]. Since the speed of sound in a polaron BEC is much higher than in an atomic BEC due to the small polaron mass, one must in fact expect the neglect of retardation effects to be even more accurate in the present case. Non-equilibrium Bogoliubov theory has moreover proven to be a reliable description of the polaron BEC [2]. Finally, our general approach is based on the well established microscopic foundation of Landau’s theory of quasiparticles and their effective interactions [45].

Recently, it has been shown that the experimental findings in Refs. [16–19] are consistent with a large decay rate of the \( \uparrow\downarrow \) bi-exciton underlying the Feshbach resonance [34]. Such a decay rate arising for instance from disorder, will likely significantly decrease the strength of the mediated interaction. In order to see the dimers discussed here, one therefore needs clean samples.

**Outlook.**—We have shown how the effective interaction between quasiparticles in exciton-polariton systems can be strong enough to support bound dimer states involving two photons. This gives rise to a new transmission line where the wave function is imprinted directly in the correlations of the transmitted light. Our results demonstrate how hybrid light-matter systems offer powerful new ways to probe many-body physics, in this case effective interactions which are a key ingredient in Landau’s
quasiparticle theory that have remained elusive so far in atom gas experiments. The possibility to engineer strong photon-photon interactions in a semiconductor microcavity moreover opens the door to realising highly non-linear optics in a solid state setting and engineering scalable optoelectronic devices such as optical gates, switches, and transistors [2, 4–13].

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[1] H. Deng, H. Haug, and Y. Yamamoto, Rev. Mod. Phys. 82, 1489 (2010). URL https://link.aps.org/doi/10.1103/RevModPhys.82.1489.
[2] I. Carusotto and C. Ciuti, Rev. Mod. Phys. 85, 299 (2013). URL https://link.aps.org/doi/10.1103/RevModPhys.85.299.
[3] G. Wang, A. Chernikov, M. M. Glazov, T. F. Heinz, X. Marie, T. Amand, and B. Urbaszek, Rev. Mod. Phys. 90, 021001 (2018). URL https://link.aps.org/doi/10.1103/RevModPhys.90.021001.
[4] T. C. H. Liew, A. V. Kavokin, and I. A. Shelykh, Phys. Rev. Lett. 101, 016402 (2008). URL https://link.aps.org/doi/10.1103/PhysRevLett.101.016402.
[5] I. A. Shelykh, R. Johne, D. D. Solnyshkov, and G. Malpuech, Phys. Rev. B 82, 153303 (2010), URL https://link.aps.org/doi/10.1103/PhysRevB.82.153303.
[6] A. Amo, T. C. H. Liew, C. Adrados, R. Houdre, E. Giacobino, A. V. Kavokin, and A. Bramati, Nature Photonics 4, 361 (2010), ISSN 1749-4893, URL https://doi.org/10.1038/nphoton.2010.79.
[7] T. Gao, P. S. Eldridge, T. C. H. Liew, S. I. Tsintzos, G. Stavrinidis, G. Deligeorgis, Z. Hatzopoulos, and P. G. Savvidis, Phys. Rev. B 85, 235102 (2012), URL https://link.aps.org/doi/10.1103/PhysRevB.85.235102.
[8] H. S. Nguyen, D. Vishnevsky, C. Sturm, D. Tanese, D. Solnyshkov, E. Galopin, A. Lemaître, I. Sagnes, A. Amo, G. Malpuech, et al., Phys. Rev. Lett. 110, 236601 (2013), URL https://link.aps.org/doi/10.1103/PhysRevLett.110.236601.
[9] T. Espinosa-Ortega and T. C. H. Liew, Phys. Rev. B 87, 195305 (2013), URL https://link.aps.org/doi/10.1103/PhysRevB.87.195305.
[10] A. V. Zasedatelev, A. V. Baranikov, D. Urbonas, F. Scalfirimuto, U. Scherf, T. Stäferle, R. F. Mahrt, and P. G. Lagoudakis, Nature Photonics 13, 378 (2019), ISSN 1749-4893, URL https://doi.org/10.1038/s41566-019-0392-8.
[11] A. Dreismann, H. Ohadi, Y. del Valle-Inclán Redondo, R. Balili, Y. G. Rubo, S. I. Tsintzos, G. Deligeorgis, Z. Hatzopoulos, P. G. Savvidis, and J. J. Baumberg, Nature Materials 15, 1074 (2016), URL https://doi.org/10.1038/nmat4722.
[12] D. Ballarini, M. De Giorgi, E. Cancellieri, R. Houdré, E. Giacobino, R. Cingolani, A. Bramati, G. Gigli, and D. Sanvito, Nature Communications 4, 1778 (2013), URL https://doi.org/10.1038/ncomms2734.
[13] D. Sanvito and S. Kena-Cohen, Nature Materials 15, 1061 (2016), ISSN 1476-4660, URL https://doi.org/10.1038/nmat4668.
[14] G. Muñoz-Matutano, A. Wood, M. Johnsson, X. Vidal, B. Q. Baragiola, A. Reinhard, A. Lemaître, J. Bloch, A. Amo, G. Nogues, et al., Nature Materials 18, 213 (2019), URL https://doi.org/10.1038/s41563-019-0281-z.
[15] A. Delteil, T. Fink, A. Schade, S. Höfling, C. Schneider, and A. Imamoglu, Nature Materials 18, 219 (2019), URL https://doi.org/10.1038/s41563-019-0282-y.
[16] N. Takemura, S. Trebaol, M. Wouters, M. T. Portella-Oberli, and B. Deveaud, Nature Physics 10, 500 EP (2014), URL https://doi.org/10.1038/nphys2999.
[17] N. Takemura, S. Trebaol, M. Wouters, M. T. Portella-Oberli, and B. Deveaud, Phys. Rev. B 90, 195307 (2014), URL https://link.aps.org/doi/10.1103/PhysRevB.90.195307.
[18] N. Takemura, M. D. Anderson, M. Navadeh-Touphchi, D. Y. Oberli, M. T. Portella-Oberli, and B. Deveaud, Phys. Rev. B 95, 205303 (2017), URL https://link.aps.org/doi/10.1103/PhysRevB.95.205303.
[19] M. Navadeh-Touphchi, N. Takemura, M. D. Anderson, D. Y. Oberli, and M. T. Portella-Oberli, Phys. Rev. Lett. 122, 047402 (2019), URL https://link.aps.org/doi/10.1103/PhysRevLett.122.047402.
[20] I. Rosenberg, D. Liran, Y. Mazuz-Harpaz, K. West, L. Pfeiffer, and R. Rapaport, Science Advances 4 (2018), URL https://advances.sciencemag.org/content/4/10/eaaat8880.
[21] E. Togan, H.-T. Lim, S. Faelt, W. Wegscheider, and A. Imamoglu, Phys. Rev. Lett. 121, 227402 (2018), URL https://link.aps.org/doi/10.1103/PhysRevLett.121.227402.
[22] P. Knüppel, S. Ravets, M. Kroner, S. Fält, W. Wegscheider, and A. Imamoglu, Nature 572, 91 (2019), URL https://doi.org/10.1038/s41586-019-1356-3.
[23] J. Gu, V. Wätcher, L. Waldeck, D. Rhodes, A. Raja, J. C. Hone, T. F. Heinz, S. Kena-Cohen, T. Pohl, and V. M. Menon, Enhanced nonlinear interaction of polaritons via excitonic rydberg states in monolayer wse2 (2019), 1912.12544.
[24] M. Sidler, P. Back, O. Cotlet, A. Srivastava, T. Fink, M. Kroner, E. Demler, and A. Imamoglu, Nature Physics 13, 255 EP (2016), URL https://doi.org/10.1038/nphys3949.
[25] L. B. Tan, O. Cotlet, A. Berghschneider, R. Schmidt, P. Back, Y. Shimazaki, M. Kroner, and A. Imamoglu, arXiv e-prints arXiv:1903.05640 (2019), 1903.05640.
[26] L. Landau, J. Exp. Theor. Phys. 3, 920 (1957).
[27] G. Baym and C. Pethick, J. Exp. Theor. Phys. 920 (1957).
[28] D. Scalapino, Physics Reports 250, 329 (1995), ISSN 0370-1573, URL http://www.sciencedirect.com/
I. SUPPLEMENTAL MATERIAL: MEDIATED INTERACTIONS AND PHOTON BOUND STATES IN AN EXCITON-POLARITON MIXTURE

II. BOGOLIUBOV THEORY AND SINGLE QUASIPARTICLE PROPERTIES

A. Bogoliubov approach

We briefly describe the Bogoliubov theory for the exciton-polaritons forming the background. For simplicity we omit the polarisation index \( \sigma \). The Hamiltonian of the single specie (majority) is given by

\[
\hat{H} = \hat{H}_{xx} + \hat{H}_{x} + \hat{H}_{f},
\]

(S1)
where the first term comprises the light-matter interaction
\[ \hat{H}_{xc} = \sum_i \sum_k \hat{a}^\dagger_{i,k} \hat{h}_i^0 \hat{a}_{i,k}, \]
(S2)

\( \hat{a}_{i,k} (\hat{a}^\dagger_{i,k}) \) are the annihilation (creation) operators of the excitons \((i = x)\) and photons \((i = c)\), respectively, in the \(k\) momentum state, and

\[ \hat{h}_i^0 = \left( \frac{\varepsilon_x^k}{\Omega} \frac{\Omega}{\varepsilon_c^k} \right). \]
(S3)

The non-linear part of the Hamiltonian contains the interactions between co-circular excitons that come from a contact repulsive potential. It reads,
\[ \hat{H}_{xx} = \frac{1}{2} \sum_{q,k,k'} g_{\uparrow\uparrow} \left( \hat{a}^\dagger_{x,k+q} \hat{a}^\dagger_{x,k'-q} \hat{a}_{x,k} \hat{a}_{x,k} \right) \]
(S4)

where \(g_{\uparrow\uparrow}\) is the interaction between the co-circular excitons.

Additionally, we consider a driving term to the \(\uparrow\) excitons by taking the quasi-mode coupling approximation valid for high-quality mirrors.

\[ \hat{H}_f = \sum_k \Omega_f \left( \hat{a}^\dagger_{x,k} F + F^* \hat{a}_{x,k} \right), \]
(S5)

Here, \(\Omega_f\) is the quasi-mode coupling, and

\[ F(x,t) = |F_{pu}| \exp\{i (k_{pu} \cdot x - \omega_{pu} t / h)\}, \]
(S6)

is the external electric field driving the microcavity, being \(k_{pu}, \omega_{pu}\) and \(|F_{pu}|\) the momentum, frequency and amplitude of the driving field, respectively. Because we are working in the momentum representation, we take the Fourier transform of the driving

\[ F = \int dx \, F(x,t) e^{-ik \cdot x} = |F_{pu}| e^{-i\omega_{pu} t / h} g_{k,k_{pu}}. \]
(S7)

We consider that \(k_{pu} = 0\). The overall effect of this term is to adjust the chemical potential of the \(\uparrow\) excitons in the BEC by \(\omega_{pu}\), which is set to be \(\omega_{pu} = \varepsilon_{0L}^0(0)\). In the absence of exciton-exciton interactions the light-matter Hamiltonian \(H_{xc}\) can be diagonalized leading to the polariton quasiparticles

\[ \left( \begin{array}{c} \hat{a}_{x,k} \\ \hat{a}_{c,k} \end{array} \right) = \left( \begin{array}{cc} C_k & -S_k \\ S_k & C_k \end{array} \right) \left( \begin{array}{c} \hat{L}_k \\ \hat{U}_k \end{array} \right) \]
(S8)

where \(\hat{L}_k (\hat{U}_k)\) are the new bosonic lower-polariton (upper) annihilation operator, and the so-called Hopfield coefficients are \(C_k^2\) and \(S_k^2 = 1 - C_k^2\) given in the main text. After applying the Hopfield transformation and dropping the upper-polariton terms we obtain

\[ \hat{H} = \sum_k \left[ \varepsilon_{L,P}^0(k) \hat{L}_k^\dagger \hat{L}_k + \Omega_f S_k \left( \hat{L}_k^\dagger F + F^* \hat{L}_k \right) \right] + \frac{g_{\uparrow\uparrow}}{2} \sum_{q,k,k'} C_{k+q} C_{k'-q} C_k C_{\ell_{k+q}} C_{\ell_{k'-q}} \hat{L}_{k+q}^\dagger \hat{L}_{k'-q} \hat{L}_k \hat{L}_k. \]
(S9)

Now, we introduce new annihilation (and creation) operators in the rotating frame \(\hat{L}_k = L_k \exp(-i\omega_{pu} t / h)\). In this rotating frame we employ the canonical Bogoliubov approximation and write the Hamiltonian as

\[ \hat{H} = E_0 + \sum_{q,k_{pu}} E_{q_{pu}}^{\text{BEC}} \hat{\beta}_{q,\uparrow}^\dagger \hat{\beta}_{q,\downarrow}, \]
(S10)

here \(\hat{\beta}\) denote the Bogoliubov operators, and the coherence factors of the canonical transformation are given by

\[ u_q, v_q = \sqrt{\frac{1}{2} \left( \frac{\varepsilon_{L,P}^0(q) - \omega_{pu} + 2 g_{\uparrow\uparrow} \hbar \Omega_0^2 C_q^2}{E_{q_{pu}}^{\text{BEC}} \pm 1} \right), \]
(S11)
with a dispersion

\[ E_{q}^{\text{BEC}} = \sqrt{\left( \varepsilon_{0}^{L}(q) - \omega_{pu} + 2g_{11}n_{b}C_{0}^{2}C_{q}^{2} \right)^{2} - g_{11}^{2}n_{b}^{2}C_{0}^{4}C_{q}^{4}} \]  

being \( n_{b} \) the density of lower-polaritons. We note that the nonequilibrium Bogoliubov mode becomes gapped [2]. The Green’s function describing the BEC are given by

\[ G_{11}^{(\text{LP})}(q, z) = G_{22}^{(\text{LP})}(q, -z) = \left( \frac{v_{q}^{2}}{z - E_{q}^{\text{BEC}}} - \frac{u_{q}^{2}}{z + E_{q}^{\text{BEC}}} \right), \quad G_{12}^{(\text{LP})}(q, z) = G_{21}^{(\text{LP})}(q, -z) = \left( \frac{u_{q}v_{q}}{z + E_{q}^{\text{BEC}}} - \frac{v_{q}u_{q}}{z - E_{q}^{\text{BEC}}} \right). \]  

III. MEDIATED INTERACTION

B. Quasiparticle properties

For a single impurity, the quasiparticle properties can be determined following the approach in [34]

\[ G^{i}(k, \omega) = [(G_{0}^{i}(k, \omega))^{-1} - \Sigma^{i}(k, \omega)]^{-1}. \]  

The impurity propagator is the 2x2 matrix given by

\[ G^{i}(k, \omega) = \begin{pmatrix} \mathcal{G}_{xx}^{i} & \mathcal{G}_{xc}^{i} \\ \mathcal{G}_{cx}^{i} & \mathcal{G}_{cc}^{i} \end{pmatrix}, \]

where the diagonal terms \( \mathcal{G}_{xx}(k, \omega) \) and \( \mathcal{G}_{cc}(k, \omega) \), account for the excitonic and photonic parts of the impurity. The ideal propagator of the impurity is a diagonal matrix such that diag((\( G_{0}^{i}(k, \omega) \))^{-1}) = (\( \omega - \varepsilon_{k}^{i}, \omega - \varepsilon_{k}^{i} \)). We employ the scattering matrix for counter-polarised excitons, which can be written as \( T^{-1}(p, z) = \Pi(0, \varepsilon_{1}) - \Pi(p, z) \), and includes the bi-exciton energy. We calculate the self-energy as

\[ \Sigma^{i}(k, \omega) = \begin{pmatrix} \Sigma_{xx}(k, \omega) & \Omega \\ \Omega & 0 \end{pmatrix}. \]  

for the Dyson equation we identify the quasiparticle properties, that is

\[ \text{Re} G^{i}(p, \varepsilon_{k})^{-1} = 0 \]

\[ Z_{k} = \left. \frac{1}{\partial_{\omega} G^{i}(p, \omega)} \right|_{\omega = \varepsilon_{k}}. \]

While the former determines the quasiparticle branches \( \varepsilon_{k} \) in Fig. 2 (main text) the latter defines the quasiparticle residue. As shown in [34] the strong light-matter coupling and the Feshbach physics yield a rich landscape of quasiparticle features.
quasiparticle residues. Note that the photonic nature of the direct and mediated scattering is encoded then in the BEC Green’s function, scattering matrix $T$, and the exciton component of the polaron-polaritons and, as shown in Eq. (3) of the main text, the light-matter coupling leads to different results when compared to pure matter-quasiparticle scattering.

The Bethe-Salpeter equation in Eq. (4) (main text) is evaluated after a pole expansion for the $G(p,z)$ propagator and by neglecting retardation effects in Eq. S19. The pole expansion takes into account the reduction of the quasiparticle pole which is relevant for the attractive polaron that cedes spectral weight to the repulsive branch as the momentum $k$ increases in magnitude, we evaluate the poles for the $s$-wave contributions of the mediated interaction. On the other hand, due to the small mass of polaritons, the speed of sound of the BEC is of the order of $c_s = \sqrt{n_0 g_{11}/m_{LP}} \approx 10^6$ m/s, therefore, the density oscillations within the medium propagates much faster than in an atomic gas [56]. We expect an efficient exchange of Bogoliubov modes within the BEC, and our theory to be accurate when $\delta v/c_s < 1$, where $\delta v = \sqrt{(E - 2\varepsilon_{LP}(0))/m_{LP}}$, which turns to be indeed the case for the onset of the two-body body bound states. Finally, details on the Bethe-Salpeter equation can be found in [51], which has been extended to include the polaritonic features due to the strong light-matter coupling.