Mediated Interactions and Photon Bound States in an Exciton-Polariton Mixture

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The quest to realize 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 nonperturbative 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 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 dimer wave function is directly mapped onto correlations between the photons. Our findings open new routes for highly nonlinear optical materials and novel hybrid light-matter quantum systems.

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Achieving strong photon-photon interactions provides a pathway to highly nonlinear optics with a range of technological applications, and it is therefore intensely pursued using a range of different physical platforms. Exciton polaritons, in short polaritons, are hybridized states of light and excitons in semiconductors inside microcavities that have risen as a promising candidate to realize such strong interactions [1–15]. In spite of impressive experimental progress [16,17], it has, however, turned out to be difficult to make the photon-photon interaction sufficiently strong to realize these objectives. Mechanisms to increase the interaction strength include Feshbach resonances [18–21], dipolar excitons [22,23], strongly correlated electrons [24], and excitons in Rydberg states [25]. Recently, one 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 [18,21] or electrons [26–28]. 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 [29,30], leading to conventional [31] and high T_c superconductivity [32], and the fundamental interaction in particle physics [33].

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 a minority component polaritons interacting with a surrounding BEC of majority polaritons. This interaction is shown to be long range, attractive, and tunable 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 transmitted photons allowing for a direct detection.

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

$$\hat{H} = \sum_{\mathbf{k}\sigma} [\hat{x}^\dagger_{\mathbf{k}\sigma} \quad \hat{c}^\dagger_{\mathbf{k}\sigma}] \begin{bmatrix} \varepsilon^x_{\mathbf{k}} & \Omega/2 \\ \Omega/2 & \varepsilon^c_{\mathbf{k}} \end{bmatrix} \begin{bmatrix} \hat{x}_{\mathbf{k}\sigma} \\ \hat{c}_{\mathbf{k}\sigma} \end{bmatrix} + g_{\uparrow\downarrow} \sum_{\mathbf{q}} \hat{\rho}_{-\mathbf{q}\uparrow}\hat{\rho}_{\mathbf{q}\downarrow} + \frac{g_{\uparrow\uparrow}}{2} \sum_{\mathbf{q}\sigma} \hat{\rho}_{-\mathbf{q}\sigma}\hat{\rho}_{\mathbf{q}\sigma}, \qquad (1)$$

where $\hat{x}^\dagger_{\mathbf{k}\sigma}$ creates an exciton with 2D transverse momentum $\mathbf{k}$, spin σ, and kinetic energy $\varepsilon^x_{\mathbf{k}} = k^2/2m_x$. Likewise, $\hat{c}^\dagger_{\mathbf{k}\sigma}$ creates a photon with momentum $\mathbf{k}$, spin σ, and kinetic energy $\varepsilon^c_{\mathbf{k}} = k^2/2m_c + \delta$, where δ is the detuning. We have defined $\hat{\rho}_{\mathbf{q}\sigma} = \sum_{\mathbf{k}} \hat{x}^\dagger_{\mathbf{k}-\mathbf{q}\sigma}\hat{x}_{\mathbf{k}\sigma}$ and use 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 Ω, giving rise to the formation of lower

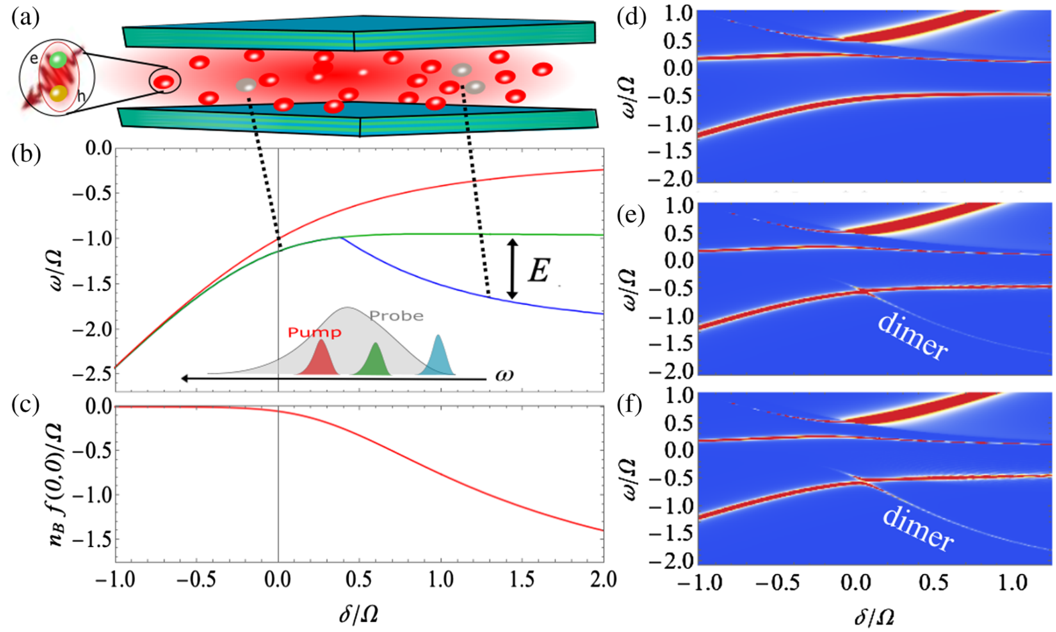

FIG. 1. (a) A pump beam creates a BEC of ↑ exciton polaritons (red balls) inside a 2D semiconductor in a microcavity. A probe beam creates ↓ quasiparticles called polaron polaritons (gray balls), which can bind via an effective interaction mediated by the BEC to form dimer states. (b) The red, green, and blue lines show energy of the ↑ polaritons, ↓ polaron polaritons, and the ↓↓ dimers respectively as a function of the detuning δ. (c) The Landau effective interaction between the ↓ polaron polaritons. The resulting photon spectral function giving the transmission of the probe beam through the semiconductor is plotted for ↓ polaron-polariton density $n = 0$, $n = 0.075n_B$, and (f) $n = 0.15n_B$ in (d),(e), and (f). This illustrates the emergence of a distinct transmission line carried by the dimer states involving two photons propagating through the BEC for $n > 0$.

and upper exciton-polariton branches with energies $\varepsilon_{\mathbf{k}} = (\varepsilon^c_{\mathbf{k}} + \varepsilon^x_{\mathbf{k}} \pm \sqrt{\delta^2_{\mathbf{k}} + \Omega^2})/2$, where $\delta_{\mathbf{k}} = \varepsilon^c_{\mathbf{k}} - \varepsilon^x_{\mathbf{k}}$.

The second line in Eq. (1) describes the interaction between excitons with opposite and parallel spins with strengths $g_{\uparrow\downarrow}$ and $g_{\uparrow\uparrow}$, 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 [34]. The interaction between ↑ and ↓ excitons supports a bound state, the biexciton, and the corresponding scattering matrix $\mathcal{T}(p)$ therefore depends strongly on momentum and energy with a pole at the biexciton energy $\varepsilon_{\uparrow\downarrow}$.

As illustrated in Fig. 1(a), we consider a BEC of exciton polaritons with density n_B and spin polarization ↑ created by a pump beam. A weaker probe beam creates a small density of exciton polaritons with spin polarization ↓, which can be regarded as impurities. Their interaction with the surrounding BEC leads to the formation of quasiparticles coined (Bose) polaron polaritons [35,36]. In Fig. 1(b), the green line shows the energy of the lowest polaron-polariton branch as a function of the detuning δ. It is below the lower polariton energy $\varepsilon_{\mathbf{k}} = (\varepsilon^c_{\mathbf{k}} + \varepsilon^x_{\mathbf{k}} - \sqrt{\delta^2_{\mathbf{k}} + \Omega^2})/2$ shown by the red line in Fig. 1(b) due to interactions with the BEC of ↑ polaritons. To calculate the energy of the polaron-polariton branch in Fig. 1(b), we have employed a diagrammatic approach where the quasiparticle energies appear as poles in a 2×2 matrix Green's function $\mathcal{G}(p) = [\mathcal{G}_0^{-1}(p) - \Sigma(p)]^{-1}$ corresponding to the Hamiltonian in Eq. (1). Here, $\mathcal{G}_0(p) = \mathrm{diag}(\omega - \varepsilon^x_{\mathbf{p}}, \omega - \varepsilon^c_{\mathbf{p}})$ is a diagonal matrix whose elements are the noninteracting exciton-photon Green's functions and the light-matter coupling is described by the off-diagonal self-energies $\Sigma_{cx} = \Sigma_{xc} = \Omega/2$. Finally, the self-energy

$$\Sigma_{xx}(p) = n_B \mathcal{C}_0^2 \mathcal{T}(p), \tag{2}$$

describes the interaction of the ↓ exciton with the ↑ exciton BEC including the Feshbach resonance nonperturbatively [35,37]. $\mathcal{C}^2_{\mathbf{q}} = (1 + \delta_{\mathbf{q}}/\sqrt{\delta^2_{\mathbf{k}} + \Omega^2})/2$ is the Hopfield coefficient of the polaritons in the BEC [39]. In the calculation of the energies in Fig. 1(b), we use realistic experimental parameters with a Rabi splitting $\Omega = 3.45$ meV, exciton mass $m_x \approx 0.16 m_e$ with m_e the electron mass, and $m_c = 10^{-4} m_x$ [17,18,40–42]. The density of the BEC is $n_B = 5 \times 10^{10}\ \mathrm{cm}^{-2}$, the direct exciton-exciton coupling $g_{\uparrow\uparrow} \approx 3\ \mu\mathrm{eV}\,\mu\mathrm{m}^2$, and the energy of the biexciton state is $\varepsilon_{\uparrow\downarrow} = 0.7$ 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 bare ↓ excitons using a nonperturbative approach that includes strong Feshbach correlations between a pair of ↑↓ excitons exactly, combined with a Bogoliubov theory generalized to the steady-state BEC at hand [3,43,44]. This gives

$$V(p,p';q) = n_B \mathcal{C}_0^2 \mathcal{C}_{\mathbf{q}}^2 \mathbf{T}^\intercal(p)\mathbf{G}^{\mathrm{LP}}(q)\mathbf{T}(p') \qquad (3)$$

for 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 $p'-q/2$. Here $\mathbf{G}^{\mathrm{LP}}(q)$ is a 2×2 matrix containing the normal and anomalous Green's functions describing sound propagation in the BEC [37]. We have defined the vector $\mathbf{T}^\intercal(p) = [\mathcal{T}(p+q/2)\mathcal{T}(p-q/2)]$ 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. For weak $\uparrow\downarrow$ interaction, Eq. (3) takes the familiar form of $V(q) = n_B \mathcal{C}_0^2 \mathcal{C}_{\mathbf{q}}^2 g_{\uparrow\downarrow}^2 \chi(q)$, which simply describes an induced interaction mediated by sound waves with a strength determined by the density-density response function $\chi(q)$ of the BEC, and with a range determined by the BEC coherence length $\propto 1/\sqrt{2m_B n_B g_{\uparrow\uparrow}}$ [45]. For stronger $\uparrow\downarrow$ interaction close to the Feshbach resonance, the mediated interaction $V(p,p';q)$ depends on both the incoming and the transferred frequency-momenta due to the lack of Galilean invariance and the finite speed of sound in the BEC.

We can now calculate the effective interaction between two $\downarrow$ polaron polaritons, which is the physically relevant quantity. As in the derivation of Landau's Fermi liquid theory [30,46], this is obtained by evaluating the mediated interaction on-shell between two polaron polaritons. Consider for concreteness the scattering between two polaron polaritons in the branch shown by the green line in Fig. 1(b). Taking both the incoming and outgoing momenta to be zero, we obtain from Eq. (3)

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

where $\varepsilon_{\mathbf{p}}$ and $Z_{\mathbf{p}}$ denote the energy and exciton residue of a polaron polariton with momentum $\mathbf{p}$. The effective interaction in Eq. (4) depends on the $\downarrow\uparrow$ scattering matrix $\mathcal{T}$ squared, reflecting that the basic mechanism is the emission and subsequent absorption of a sound mode in the BEC. 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 in a conventional BEC [45,47–49], Equation (4) contains an additional factor $2/3$ originating from the nonequilibrium nature of the BEC, as well as the residue $Z_{\mathbf{p}}$.

Figure 1(c) shows numerical results for the effective interaction $f(\mathbf{0},\mathbf{0})$ between two polaron polaritons as a function of the detuning δ. We see that the interaction is attractive and increases in strength with the detuning, becoming of the order $f(\mathbf{0},\mathbf{0}) \sim \Omega/n_B$, when $\delta \gtrsim \Omega/2$. Assuming a polaron-polariton density n not too small compared to n_B, this corresponds to a large mean-field shift $nf(\mathbf{0},\mathbf{0})$ of the order of $n/n_B\Omega$. The mechanism for this strong interaction is twofold. First, the exciton component, which is the one that interacts, increases with increasing δ. Second, the energy of the attractive lower polaron-polariton approaches that of the biexciton with increasing δ giving rise to a strong $\uparrow\downarrow$ exciton scattering and thereby a large effective interaction.

Dimer states.—The strong effective interaction between two $\downarrow$ polaron polaritons combined with its long range nature determined by the coherence length of the BEC, and the 2D geometry raises the intriguing possibility of bound states consisting of two $\downarrow$ polaron polaritons. To capture the presence of such dimers, we have to account for the strong correlations arising as a consequence of the repeated scattering between two $\downarrow$ excitons via the exchange of sound modes in the BEC. This is done via the Bethe-Salpeter equation (BSE) for the scattering matrix Γ between a pair of $\downarrow$ excitons, which in the ladder approximation reads [50]

$$\Gamma(k_1,k_2;k_1-k_3) = V(k_1,k_2;k_1-k_3) - \sum_q V(k_1,k_2;q) \times G(k_1-q)G(k_2+q)\Gamma(k_1-q,k_2+q;k_1-q-k_3). \qquad (5)$$

Here, $G(k)$ is the Green's function of the $\downarrow$ excitons coupled to light and interacting with the BEC.

It is very complicated to solve the BSE taking into account the full momentum-energy dependence of the interaction $V(k_1,k_2;k_3)$, and we therefore make two approximations. First, we employ a pole expansion of $G(k)$, which corresponds to assuming that the $\downarrow$ excitons exclusively exist in the polaron-polariton state when they are unbound. Second, we neglect retardation effects by setting the frequency to zero in the mediated interaction $V(k_1,k_2;k_3)$. Even when both approximations are performed, the numerical calculation is still quite involved. Details are given in the Supplemental Material [37].

The blue line in Fig. 1(b) shows the binding energy E of the dimer relative to the polaron polariton obtained from solving $\Gamma^{-1}(E) = 0$ for zero center of mass momentum. Remarkably, we see that the mediated interaction indeed is strong enough to bind two polaron polaritons into a dimer state beyond a critical detuning $\delta \gtrsim 0.5\Omega$. The binding energy of the dimer increases with δ reflecting the increasing attraction in agreement with Fig. 1(c), and it becomes comparable to the energy shift of the polaron polariton with respect to the bare polariton. Since the polaron polaritons are partly photons these dimers in turn correspond to bound states of two photons. The existence

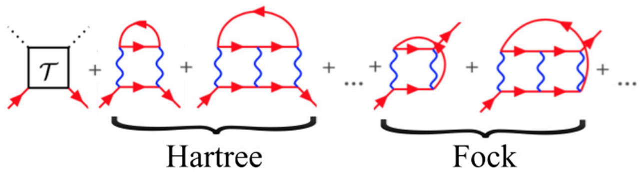

FIG. 2. The ↓ exciton self-energy with the first term describing scattering with the BEC giving Eq. (2) and the next two terms describing the interaction with other ↓ polaron polaritons via the mediated interaction giving Eq. (6). Red lines are the ↓ exciton propagator, dashed lines are ↑ polaritons in the BEC, the box is the $\mathcal{T}$ matrix, and the wavy line is the mediated interaction in Eq. (3). The first order term in the mediated interaction is included in the $\mathcal{T}$ matrix.

of an mediated photon-photon interaction strong enough to support bound states is a main result of this work.

Note that in a vacuum there is always a bound state for any attractive interaction between two particles [51]. The reason the bound state only exists beyond a critical detuning in the present case is due to many-body effects, which alter the one particle dispersion into that of a polaron polariton and make the mediated interaction nonlocal, i.e., depending on all momenta [52].

Light transmission.—We now show that the dimer state gives rise to a distinct line of transmitted light. To do this, we include the correlations leading to the formation of the dimer in the self-energy as

$$\Sigma_{\rm in}(p) = nZ_p^2[\Gamma(p,0;0) + \Gamma(p,0;-p)]. \qquad (6)$$

Equation (6) gives the energy shift of the polaron polariton due to its interactions with other polaron polaritons of density n in the Hartree-Fock approximation. We have replaced the induced interaction with the scattering matrix Γ obtained from the BSE to include strong coupling effects [37,50]. The Feynman diagrams corresponding to Eq. (6) are shown in Fig. 2. Adding $\Sigma_{\rm in}(p)$ to Eq. (2) includes dimer formation in our many-body theory for the polaron polaritons.

In Figs. 1(d)–1(f), we plot the ↓ photon spectral function as a function of the detuning δ. There are several polaron-polariton branches typical of the interplay between strong interactions and light coupling [26,28,35,36]. The lowest branch corresponds to the green line in Fig. 1(b). Compared to the spectrum for vanishing polaron-polariton density shown in Fig. 1(d), the spectra for the polaron-polariton densities $n = 0.075n_B$ (e) and $n = 0.15n_B$ (f) exhibit one qualitative new feature: A dimer line has emerged, which 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. Comparing Figs. 1(e) and 1(f), we see that the strength of this dimer line increases with n because there are then more polaron polaritons in the BEC to form dimers with. The onset of a dimer state is moreover accompanied by an avoided crossing between the lower polaron-polariton branch and the dimer branch, which leads to significant energy shifts compared to the case of no dimer states.

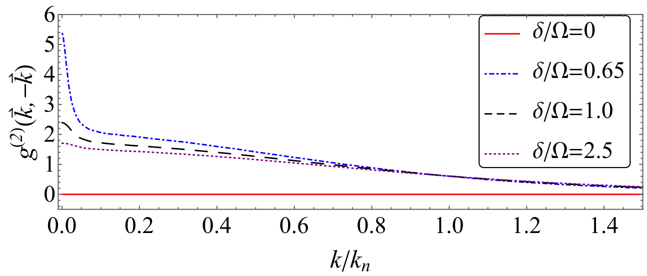

FIG. 3. Correlations between transmitted photons with opposite transverse momenta as parametrized by $g_2(\mathbf{p}, -\mathbf{p})$ for different detunings.

Photon correlations.—We finally show that the wave function of the dimer is imprinted directly on the transmitted light correlations. Figure 3 plots the correlation function $g_2(\mathbf{p}, -\mathbf{p}) = \langle a^\dagger_{\mathbf{p}} a_{\mathbf{p}} a^\dagger_{-\mathbf{p}} a_{-\mathbf{p}}\rangle - \langle a^\dagger_{\mathbf{p}} a_{\mathbf{p}}\rangle\langle a^\dagger_{-\mathbf{p}} a_{-\mathbf{p}}\rangle$ for different detunings δ, where, $a^\dagger_{\mathbf{p}}$ creates a polaron polariton with momentum $\mathbf{p}$. It is calculated using the wave function $|\Phi\rangle = \sum_{\mathbf{p}>0}\phi(\mathbf{p}) a^\dagger_{\mathbf{p}} a^\dagger_{-\mathbf{p}}|0\rangle$ of the dimer as $g_2(\mathbf{p}, -\mathbf{p}) = |\phi(\mathbf{p})|^2 - |\phi(\mathbf{p})|^4$, where $|0\rangle$ is the vacuum state. Here, $\phi(\mathbf{p})$ is obtained from the BSE by mapping it onto an effective Schrödinger equation [37,52]. We see that g_2 indeed is nonzero when there is a bound state. The correlations moreover extend to increasingly high momenta with increasing δ, which directly reflects the decreasing spatial size of the dimer wave function with increasing binding energy.

Discussion.—Since we are considering a strongly correlated hybrid light-matter system, it is worth discussing our approach. First, the ladder approximation describing the formation of polaron polaritons is surprisingly accurate for the analogous problem of atomic polaron formation [53–62]. A similar theory for the mediated interaction between impurities and dimer formation in an atomic BEC has, moreover, been shown to be remarkably accurate even for strong interactions when benchmarked against Monte Carlo calculations [48,52]. Nonequilibrium Bogoliubov theory is known to be a reliable description of the polariton BEC [3]. Since our results are based on the existence of a linear sound spectrum in the medium, we expect them to be robust towards fragmentation of the BEC [63,64]. The intrinsic decay due to photon leakage out of the cavity will not significantly affect our results, as long as the resulting linewidths are small compared to their separation. Finally, our approach is based on the well-established microscopic foundation of Landau's theory of effective interactions between quasiparticles [46].

Recently, it has been shown that the experimental findings in Refs. [18–21] are consistent with a fast decay of the ↑↓ biexciton underlying the Feshbach resonance [35]. Such a decay will likely decrease the strength of the mediated interaction. In order to see the dimers discussed here, one therefore needs clean samples.

The energy shift of the lower polaron-polariton line due to the avoided crossing with the dimer state should be readily detectable. Indeed, the energy shift is $\sim 0.25\Omega = 0.86$ meV for $\delta/\Omega \simeq 0.18$, where the dimer state emerges for the parameters in Fig. 1(f). The spectral weight of the dimer line is moreover ~20% of that of the lowest polaron-polariton line in this regime [37], which is well within present experimental resolution [27,28]. This, combined with the fact that it can be switched on or off by the pump beam, make the prospects for its observation very good. One could 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 clearly reflecting its nonlinear nature. Finally, Fig. 3 shows that measuring the photon-photon correlations in the transmitted light will provide a smoking gun detection of the bound state wave function.

Outlook.—We have shown how the effective interaction between polaron polaritons in a semiconductor can be strong enough to support 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. The possibility to engineer mediated strong photon-photon interactions in a semiconductor microcavity moreover opens the door to realizing highly nonlinear optics in a solid state setting and engineering scalable optoelectronic devices such as gates, switches, and transistors [3,5–9, 11–15]. We finally note that strongly interacting polaron polaritons can also be realized in atomic systems [65–67].

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