Dipolar polaritons squeezed at unitarity

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Interaction of dipolar polaritons can be efficiently tuned by means of a shape resonance in their excitonic component. Provided the resonance width is large, a squeezed population of strongly interacting polaritons may persist on the repulsive side of the resonance. The derived analytical expression for the polariton coupling constant reveals an excellent agreement with the puzzling experimental observations [I. Rosenberg et al., Sci. Adv. 4, 8880 (2018)]. Our arguments provide a new direction for the quest of interactions in quantum photonics.

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The commonly adopted strategy to introduce interactions into the quantum optics of semiconductors is tailoring the non-linearity due to excitonic transitions [1]. In the regime of strong light-matter coupling the macroscopic population of the cavity mode is efficiently transferred into the exciton field, which can be regarded as a gas of bosonic quasiparticles [2–4]. In particular, the blueshift of the polariton dispersion is governed by low-energy s-wave collisions between the pairs of excitons [2]. This naturally refers to ultra-cold atomic systems, where enhancement of interactions has been demonstrated by working with species having dipole moments [5], Rydberg excitations [6] and using the technique of Feshbach resonance [7]. The latter provides a possibility to tune the scattering length from positive to negative values through the unitary limit by adjusting the position of the scattering threshold with respect to a bound state.

On the technological side, exceptional excitonic properties are found in atomically thin heterostructures of transition metal dichalcogenides (TMD’s) [8]. The atom-like Lennard-Jones interaction between excitons has been demonstrated in these materials [9]. Such interaction naturally admits a bound state and, indeed, biexcitons have recently been observed in several types of monolayers [10]. A fundamental difference from the atomic clouds is, however, a purely two-dimensional (2D) character of the exciton translational motion. The interactions in a 2D ultra-cold gas are generically weak due to the properties of 2D kinematics. Thus, in contrast to three dimensions, quantum scattering off a weakly-bound state has a vanishingly small amplitude [11]. At sufficiently low exciton densities these arguments apply also for semiconductor quantum wells (QW’s).

As was proposed by the author [12], a 2D analog of the Feshbach resonance may be realized with dipolar excitons formed of electrons and holes residing in spatially separated layers. The dipolar repulsion introduces a potential barrier between the outer continuum and the bound state (biexciton), which enables a quasi-discrete level with tunable energy ε and lifetime ℏ/3. Both parameters can be controlled by changing the distance d between the layers. The attractive side of such resonance was theoretically explored in the context of roton-maxon excitations and supersolidity in dipolar Bose-Einstein condensates (BEC’s) [13, 14]. On the repulsive side, the equilibrium ground state is a condensate of biexcitons, distinguished from the exciton condensate by suppressed coherence of the photoluminescence (PL) and a gapped excitation spectrum [12]. These predictions hold for a wide variety of bilayer structures, where the exciton lifetime is sufficiently long to establish a thermodynamic equilibrium. Thus, the numerical calculations of the exciton interaction potential in coupled QW’s [15] suggest that the shape resonance may be responsible for the formation of a fragmented-condensate solid of excitons [13, 14, 16, 17].

Several groups have recently reported an increase of the polariton interaction due to the dipolar moment in the excitonic component [18, 20]. The results presented in Ref. [19] are particularly compelling: a factor of 200 enhancement of the dipolar polariton interaction strength as compared to unpolarized polaritons have been detected. Dipolar repulsion alone cannot explain such tremendous blueshift of the polariton PL. The mystery is deepened by very low values of polariton densities at which the experiment was done.

Motivated by these experimental observations, the Letter presents a phenomenological model of resonantly paired dipolar polaritons. In contrast to dipolar excitons, microcavity polaritons are far from the thermodynamic equilibrium, their statistics being closer to lasers rather than to atomic BEC’s [21]. This makes possible existence of a metastable polariton population on the repulsive side of the shape resonance. Coupling to a transient bipolaron mode in this case yields divergent behaviour of the 2D effective interaction, akin to the unitary limit in three-dimensional atomic clouds. The derived analytical expression for the interaction enhancement factor as a function of the polariton dipole moment and density shows an excellent agreement with the experimental results of Ref. [19]. Another interesting prediction of our theory is that the many-body polariton states become squeezed by the resonance. This could be verified in current experiments by examining the statistics of emitted photons.
Let us discuss the relevant timescales of the problem. First, we shall assume that the polaritons do not relax to the paired state, which is the equilibrium ground state when \( \varepsilon \) is below the scattering threshold. Second, the width of the resonance \( \beta \) must be sufficiently large for polaritons could feel the interior of the barrier during their lifetime \( \tau \). Hence, we let

\[
h/\beta \ll h/\varepsilon \ll \tau \ll \tau_k,
\]

where \( \tau_k \) is the thermalization time.

Furthermore, for the sake of simplicity we consider a single polariton branch characterized by the effective mass \( m \). Analysis of a possible departure from this approximation will be given elsewhere. The system is a mixture of two polariton flavours \( c_\sigma \) with \( \sigma = (\uparrow, \downarrow) \) and their bipolaronic pairs \( C \). In practice, "\( \uparrow \)" and "\( \downarrow \)" typically correspond to left- and right-circularly polarized photons.

The many-body Hamiltonian reads

\[
\hat{H} = \sum_{p,\sigma} \left( \frac{\hbar^2 p^2}{2m} + E_0 \right) \hat{c}_{\sigma,p} \hat{c}_{\sigma,p} + \sum_k \left( \frac{\hbar^2 k^2}{4m} + 2E_0 + \varepsilon \right) \hat{C}_{k} \hat{C}_{k} \quad + \frac{g}{2S} \sum_{p_1,p_2,q,\sigma} \hat{c}_{\sigma,p_1+q} \hat{c}_{\sigma,p_2-q} \hat{c}_{\sigma,p_1} \hat{c}_{\sigma,p_2}
\]

The condition (1) provides a physical meaning to the interaction of polaritons with opposite spins by converting them into the bipolariton mode and vice versa. The square-root prefactor is constructed in such a way as to reproduce the low-energy 2D scattering amplitude for two particles in vacuum [12].

By using the standard commutation relations for bosons, one obtains the following set of Heisenberg equations of motion

\[
\begin{align}
\frac{i\hbar}{\mu_\sigma} \frac{d\hat{c}_{\sigma,p}}{dt} &= \left( \frac{\hbar^2 p^2}{2m} + E_0 + \mu_\sigma \right) \hat{c}_{\sigma,p} + \sqrt{\frac{\hbar^2 \beta}{2\pi m S}} \sum_k \hat{c}^\dagger_{\sigma',\sigma,k} \hat{C}_{k+p} \\
\frac{i\hbar}{\mu} \frac{d\hat{C}_{k}}{dt} &= \left( \frac{\hbar^2 k^2}{4m} + 2E_0 + \varepsilon \right) \hat{C}_{k} + \sqrt{\frac{\hbar^2 \beta}{2\pi m S}} \sum_p \hat{c}^\dagger_{\uparrow,-p+\frac{1}{2}} \hat{c}_{\downarrow,p+\frac{1}{2}}
\end{align}
\]

where \( \mu_\sigma = \frac{g}{S} \sum_q |c_{\sigma,q}|^2 = gn_\sigma \),

with \( n_\sigma \) being the polariton densities in each component. By introducing the slowly-varying amplitudes

\[
\hat{c}_{\sigma,p} = \hat{c}_{\sigma,p} e^{-i(\frac{\hbar^2 p^2}{2m} + E_0 + \mu_\sigma)t/\hbar},
\]

we notice existence of a stationary (\( d\hat{c}_{\sigma,p}/dt = 0 \)) solution of Eq. (3b) in the form

\[
\hat{C}_{k} = \frac{\sqrt{\frac{\hbar^2 \beta}{2\pi m S}}}{\mu_\uparrow + \mu_\downarrow - \varepsilon' \sum_p} \sum_p \hat{c}^\dagger_{\uparrow,-p+\frac{1}{2}} \hat{c}_{\downarrow,p+\frac{1}{2}},
\]

where the motion of the bipolariton mode is reduced to that of a pair of polaritons with opposite spins. Here \( \varepsilon' = \varepsilon - \varepsilon_k \) with

\[
\varepsilon_k = \frac{\sum_p \frac{\hbar^2 p^2}{m} < \hat{c}^\dagger_{\uparrow,-p+\frac{1}{2}} \hat{c}_{\downarrow,p+\frac{1}{2}} >}{\sum_p < \hat{c}^\dagger_{\uparrow,-p+\frac{1}{2}} \hat{c}_{\downarrow,p+\frac{1}{2}} >}
\]

being the kinetic energy of the relative motion in the pair.

The condition (1) provides a physical meaning to the solution [6]. The objects \( \hat{C}_{k} \) should be regarded as auxiliary fields describing onset of pair correlations between the polaritons, rather than new (quasi)particles. Indeed, substituting (6) into the last term of the Hamiltonian [2],
one obtains an effective model

\[ \hat{H}' = \sum_{p, \sigma} \left( \frac{\hbar^2 p^2}{2m} + E_0 \right) \hat{c}_{\sigma, p}^{\dagger} \hat{c}_{\sigma, p} + \right. \]

\[ \left. \frac{1}{2S} \sum_{p_1, p_2, \sigma, \sigma'} \hat{c}_{\sigma, p_1}^{\dagger} \hat{c}_{\sigma, p_2}^{\dagger} q_{\sigma' \sigma, p_2} \hat{c}_{\sigma', p_1} \hat{c}_{\sigma', p_2}, \right \] \tag{8} \]

with \( g_{\uparrow \uparrow} = g_{\downarrow \downarrow} = g \) and

\[ g_{\uparrow \downarrow} = \frac{\hbar^2}{2\pi m} \left( \mu_{\uparrow} + \mu_{\downarrow} - \varepsilon' \right). \tag{9} \]

The bipolariton part (which we have conveniently omitted) in this picture is completely decoupled from the polariton dynamics, the pair correlations manifesting themselves as a resonant inter-component interaction. This interaction becomes increasingly strong as the background polariton blueshift approaches the renormalized energy of the discrete level. Though in the present work we are primarily concerned with the case \( \mu_{\uparrow} + \mu_{\downarrow} > \varepsilon' \), the formula \( \varepsilon_{\text{gr}} \) can be used on the attractive side \( \mu_{\uparrow} + \mu_{\downarrow} < \varepsilon' \) as well.

In the experiment [19] a laser pulse generates a population of the heavy-hole dipolar polaritons in a waveguide cavity with wide GaAs QW’s. The output signal is registered at the distance \( l \sim \nu_{gr} \tau \) from the excitation spot. The polariton group velocity \( \nu_{gr} \) is several orders of magnitude larger than the corresponding quantity for bare excitons. A possible reservoir of excitons can therefore be safely ignored in our consideration.

Assume that at \( t = 0 \) the polariton gas is a coherent balanced mixture, \( n_{\uparrow} = n_{\downarrow} = n = N/S \), occupying a single-particle state with some definite \( k_0 \) on the dispersion curve:

\[ \hat{c}_{\sigma, k_0}(0)|\psi\rangle = \sqrt{N}|\psi\rangle \]

\[ \hat{c}_{\sigma, k \neq k_0}(0)|\psi\rangle = 0. \tag{10} \]

The bipolariton part of the many-body wavefunction \( |\psi\rangle \) is initially in the vacuum state:

\[ \hat{C}_{2k_0}(0)|\psi\rangle = 0. \tag{11} \]

By substituting the slowly varying \( \epsilon \)-numbers [see the definition (5)] \( c_{\sigma, k_0} = \rho_{\sigma} e^{i\phi_{\sigma}} \) and \( \hat{C}_{2k_0} = \hat{\rho}_e e^{i\phi} \) into Eqs. (3), and omitting the terms scaling as \( \sqrt{\epsilon/\beta} \), one can find

\[ \phi(t) = \phi_{\uparrow}(t) + \phi_{\downarrow}(t) \pm \pi/2 \]

\[ \phi_{\sigma}(t) = \phi_{\sigma}(0) \] \tag{12}

and

\[ \rho_{\sigma}(t) = \sqrt{N} \cosh^{-1}(t/\tau_0) \]

\[ \rho(t) = \sqrt{N} \tanh(t/\tau_0). \tag{13} \]

Eq. (13) shows that on the characteristic time scale

\[ \tau_0 = \sqrt{\frac{2\pi m}{\beta n}} \ll \tau \] \tag{14}

the coherent mixture is entirely converted into the paired state \( \langle \bar{\phi} \rangle \). According to (8) and (9), the modified polariton blueshift is given by

\[ \mu'_{\sigma} = g n + \frac{\hbar^2 n}{2\pi m} \frac{\beta}{(2ng - \varepsilon)}. \tag{15} \]

The width of the resonance changes from 0 to \( \infty \) (the latter describing the ultimate case where the level washes out) as the exciton dipole moment is tuned from \( d \ll d_c \) to \( d \gg d_c \), where \( d_c \) is the critical value at which the true bound state disappears. We shall assume \( 0 \leq d \lesssim d_c \) and take \( \beta(d) = Bd \). The corresponding dependence for the position of the level has the form (12) \( \varepsilon(d) = \mathcal{E}(d - d_c) \).

The authors of Ref. [19] plot the quantity \( \eta(d, n) = \mu'_{\sigma}/\mu_{\sigma} - 1 \), which they call the ”interaction enhancement factor”, as a function of the dipole moment \( d \) and density \( n \). Substituting the above relations for \( \beta(d) \) and \( \varepsilon(d) \) into Eq. (15), we obtain

\[ \eta(d, n) = \frac{\hbar^2}{2\pi mg} \frac{Bd}{[2ng - \mathcal{E}(d - d_c)]}. \tag{16} \]
The experimental data of Ref. [19] fitted by the analytical expression [16] are shown in Fig. 1. We take $g = 20 \mu eV \times \mu m^2$, $d_c = 10 \mu m$ [22] and $m = 10^{-3} m_X$, where $m_X$ is the effective mass of the heavy-hole exciton in a GaAs QW. From the fitting we find $B = 400 \mu eV \times \mu m^2$ and $\xi = 1.2 \mu eV \times \mu m^{-1}$, which at $d = 1 \mu m$ yields $\hbar/\beta \approx 1$ ps and $|\varepsilon| \approx 10 \mu eV$. With the experimentally achieved polariton lifetime $\tau \approx 100$ ps the requirement $|\varepsilon| \approx 10 \mu eV$ is well fulfilled [23], which justifies our approach a posteriori.

Interestingly, the strong correlations in the paired state [9] squeeze the polariton wavefunctions. To illustrate this point, consider again the situation close to the experimental one discussed above, where one starts from a coherent state [10] for polaritons and a vacuum state [11] for their pairs. Introduce rotated quadratures
\[ \hat{x}_\sigma = \frac{1}{2} (\xi_{\sigma, k_0} e^{-i \phi_\sigma} + \xi_{\sigma, k_0} e^{i \phi_\sigma}) \]
\[ \hat{y}_\sigma = \frac{1}{2} (\xi_{\sigma, k_0} e^{-i \phi_\sigma} - \xi_{\sigma, k_0} e^{i \phi_\sigma}) \] (17)

and
\[ \hat{X} = \frac{1}{2} (\hat{C}_{2k_0} e^{-i \phi} + \hat{C}_{-2k_0} e^{i \phi}) \]
\[ \hat{Y} = \frac{1}{2} (\hat{C}_{2k_0} e^{-i \phi} - \hat{C}_{-2k_0} e^{i \phi}). \] (18)

Write $\hat{x}_\sigma = x_\sigma + \delta \hat{x}_\sigma$ and the same for $\hat{y}_\sigma$, $\hat{X}$, $\hat{Y}$. The linearized equations of motion for the quadrature fluctuations read
\[
\frac{d}{dt} \delta \hat{x}_{\uparrow, \downarrow} = \pm \sqrt{\frac{\beta}{2\pi m_S}} \left( \rho_{\uparrow, \downarrow} \delta \hat{X} + \rho \delta \hat{x}_{\downarrow, \uparrow} \right) \\
\frac{d}{dt} \delta \hat{y}_{\uparrow, \downarrow} = \pm \sqrt{\frac{\beta}{2\pi m_S}} \left( \rho_{\uparrow, \downarrow} \delta \hat{Y} - \rho \delta \hat{y}_{\downarrow, \uparrow} \right) \\
\frac{d}{dt} \delta \hat{X} = \mp \sqrt{\frac{\beta}{2\pi m_S}} \left( \rho_\uparrow \delta \hat{x}_\downarrow + \rho_\downarrow \delta \hat{x}_\uparrow \right) \\
\frac{d}{dt} \delta \hat{Y} = \mp \sqrt{\frac{\beta}{2\pi m_S}} \left( \rho_\uparrow \delta \hat{y}_\downarrow + \rho_\downarrow \delta \hat{y}_\uparrow \right), \] (19)

where the sign “+” or “−” corresponds to the two possible choices of the phase shift in Eq. (12), and $\rho$, $\rho_\sigma$ are given by [13]. At $t = 0$ one can use Eqs. (17) and (10) to find $\langle \delta \hat{x}_{\uparrow}^2(0) \rangle = 1/4$ and $\langle \delta \hat{y}_{\uparrow}^2(0) \rangle = 1/4$, the well-known property of a coherent state [24]. In contrast, at $\tau_0 \ll t \ll \tau$, where $\tau_0$ is given by Eq. (14), one can substitute $\rho_\sigma = 0$ and $\rho = \sqrt{N}$ into the first pair of Eqs. (19) to obtain
\[ \langle \delta \hat{x}_{\uparrow}^2(t) \rangle \sim e^{\pm t/\tau_0} \]
\[ \langle \delta \hat{y}_{\uparrow}^2(t) \rangle \sim e^{\mp t/\tau_0}, \] (20)

showing that the polaritons exhibit 100% squeezing in either of the two quadratures at the output.

The requirement [11] has allowed us to neglect the dissipation and make our arguments particularly transparent. Pair-breaking events due to leakage of the single photons from the cavity (e.g., through the grating out-coupler in the experiment [19]) result in loss of correlations and, at a first glance, would reduce the degree of squeezing. In practice, however, this reduction may be fully compensated by the noise of the external vacuum (see Ref. [25]), which restores the significance of the result [20].

Our last remark concerns the choice of the sign in Eq. (12). Under the condition $|\varepsilon| \approx |\varepsilon_0|$, the Josephson coupling of the polariton states to the bound state stabilizes a definite phase relation during the signal propagation. The initial configuration is, however, chosen stochastically and may vary from one laser pulse to another. This circumstance should be taken into account when verifying the prediction [20] experimentally.

To conclude, we have explained the anomalously large enhancement of repulsive interactions in a system of dipolar polaritons reported in Ref. [19]. The proposed model is based on the physics of a bound state separated from the outer continuum by a potential barrier. Our results apply to a wide variety of 2D semiconductor heterostructures, including the atomically thin layers of TMD’s. An intriguing prediction of our theory is that the resonantly paired polaritons represent an efficient source of squeezed radiation. This might be readily verified by examining the statistics of emitted photons with the balanced homodyne detection [26]. The idea of using the shape resonance to produce strong pair correlations and squeezing at ultra-low polariton densities opens wide perspectives for future research and applications. Thus, an interesting new direction would be application of the physics discussed in this work to the recently established field of topological polaritons [27, 28].

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[1] I. Carusotto and C. Ciuti, Rev. Mod. Phys. 85, 299 (2013).
[2] L. V. Keldysh and A. N. Kozlov, Sov. Phys. JETP 27, 521 (1968).
[3] E. Hanamura and H. Haug, Phys. Rep. 33, 209 (1977).
[4] P. Remucci, T. Amand, X. Marie, P. Senellart, J. Bloch, B. Servin, and K. V. Kavokin Phys. Rev. B 72, 075317 (2005).
[5] T. Lahaye, C. Menotti, L. Santos, M. Lewenstein, and T. Pfau, Rep. Prog. Phys. 72, 126401 (2009).
[6] R. Low, H. Weimer, J. Nipper, J. B. Balsewski, B. Butcher, H. P. Buchler, and T. Pfau, J. Phys. B At. Mol. Opt. Phys. 45, 113001 (2012).
[7] C. Chin, R. Grimm, P. Julienne, and E. Tiesinga, Rev. Mod. Phys. 82, 1225 (2010).
[8] Gang Wang, Alexey Chernikov, Mikhail M. Glazov, Tony F. Heinz, Xavier Marie, Thierry Amand, and Bernhard Urbaszek, Rev. Mod. Phys. 90, 021001 (2018).
[9] E. J. Sie, A. Steinhoff, C. Gies, C. H. Lui, Q. Ma, M. Rosner, G. Schonhoff, F. Jahne, T. O. Wehling, Y.-H. Lee, J. Kong, P. Jarillo-Herrero, and N. Gedik, Nano Letters
[10] Y. You, X.-X. Zhang, T. C. Berkelbach, M. S. Hybertsen, D. R. Reichman, and T. F. Heinz, Nature Physics 11, 477 (2015); Z. Ye, L. Waldecker, E. Y. Ma, D. Rhodes, A. Antony, B. Kim, X.-X. Zhang, M. Deng, Y. Jiang, Z. Lu, D. Smirnov, K. Watanabe, T. Taniguchi, J. Hone, T. F. Heinz, Nat. Commun. 9, 3718 (2018); Z. Li, T. Wang, Z. Lu, C. Jin, Y. Chen, Y. Meng, Z. Lian, T. Taniguchi, K. Watanabe, S. Zhang, D. Smirnov, S.-F. Shi, Nat. Commun. 9, 3719 (2018).

[11] L. D. Landau and E. M. Lifshitz, Quantum Mechanics (Pergamon Press, Oxford, 1969).

[12] S. V. Andreev, Phys. Rev. B 94, 140501(R) (2016).

[13] S. V. Andreev, Phys. Rev. B 92, 041117(R) (2015).

[14] S. V. Andreev, Phys. Rev. B 95, 184519 (2017).

[15] C. Schindler and R. Zimmermann, Phys. Rev. B 78, 045313 (2008).

[16] S. V. Andreev, Phys. Rev. Lett. 110, 146401 (2013).

[17] S. V. Andreev, A. A. Varlamov and A. V. Kavokin, Phys. Rev. Lett. 112, 036401 (2014).

[18] S. I. Tsintzos, A. Tzimis, G. Stavrinidis, A. Trifonov, Z. Hatzopoulos, J. J. Baumberg, H. Ohadi, and P. G. Savvidis, Phys. Rev. Lett. 121, 037401 (2018).

[19] I. Rosenberg, D. Liran, Y. Mazuz-Harpaz, K. West, L. Pfeiffer, and R. Rapaport, Sci. Adv. 4, 8880 (2018).

[20] Emre Togan, Hyang-Tag Lim, Stefan Faelt, Werner Wegscheider, and Atac Imamoglu, Phys. Rev. Lett. 121, 227402 (2018).

[21] L. V. Butov and A. V. Kavokin, Nat. Photon. 6, 2 (2012); M. Klaas, E. Schlottmann, H. Flayac, F. P. Laussy, F. Gericke, M. Schmidt, M. v. Helversen, J. Beyer, S. Brodbeck, H. Suchomel, S. Hofling, S. Reitzenstein, and C. Schneider, Phys. Rev. Lett. 121, 047401 (2018).

[22] A. D. Meyerholen and M. M. Fogler, Phys. Rev. B 78, 235307 (2008); I. V. Bondarev and M. R. Vladimirova, Phys. Rev. B 97, 165419 (2018).

[23] Note, however, that the reported values of $\tau$ approach the lifetime which was shown to be sufficient for polaritons with moderate interactions to reach the equilibrium statistical distribution, see Yongbao Sun, Patrick Wen, Yo-seob Yoon, Gangqiang Liu, Mark Steger, Loren N. Pfeiffer, Ken West, David W. Snoke, and Keith A. Nelson, Phys. Rev. Lett. 118, 016602 (2017).

[24] M. O. Scully and M. S. Zubairy, Quantum Optics (Cambridge University Press, 1997).

[25] B. Yurke, Phys. Rev. A 29, 408 (1984).

[26] G. Breitenbach, S. Schiller, and J. Mlynek, Nature 387, 471 (1997).

[27] A. V. Nalitov, D. D. Solnyshkov, and G. Malpuech, Phys. Rev. Lett. 114, 116401 (2015).

[28] Tomoki Ozawa, Hannah M. Price, Alberto Amo, Nathan Goldman, Mohammad Hafezi, Ling Lu, Mikael C. Rechtsman, David Schuster, Jonathan Simon, Oded Zilberberg, and Iacopo Carusotto, Rev. Mod. Phys. 91, 015006 (2019).