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. We derive an analytical expression for the polariton coupling constant, and we estimate the degree of squeezing of the emitted light. The squeezing may approach 100% in typical experimental conditions. Our arguments hold promises for implementation of strong correlations in quantum photonics.

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The commonly adopted strategy to introduce interactions into the quantum optics of semiconductors is tailoring the nonlinearity 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 [2–5]. 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 ultracold atomic systems, where enhancement of interactions has been demonstrated by working with species having dipole moments [6], Rydberg excitations [7] and using the technique of Feshbach resonance [8]. 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) [9]. The atomlike Lennard-Jones interaction between excitons has been demonstrated in these materials [10]. Such interaction naturally admits a bound state, and indeed biexcitons have recently been observed in several types of monolayers [11–13]. 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 ultracold 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 [14]. At sufficiently low exciton densities, these arguments apply also for semiconductor quantum wells (QWs).

As was proposed by the author [15], a 2D analog of the Feshbach resonance may be realized with dipolar excitons formed out 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 quasidiscrete level with tunable energy and lifetime. 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 (BECs) [16,17]. 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 [15]. 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 QWs [18] suggest that the shape resonance may be responsible for the formation of a fragmented-condensate solid of excitons [16,17,19,20].

Several groups have recently reported an increase of the polariton interaction due to the dipolar moment in the excitonic component [21–23]. The results presented in Ref. [22] are particularly compelling: A factor of 200 enhancement of the dipolar polariton interaction strength as compared to unpolarized polaritons has been detected. Dipolar repulsion alone cannot explain such a 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 paper 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 BECs [24,25]. This enables the existence of a metastable polariton population on the repulsive side of the shape resonance. Coupling to a transient bipolariton mode in this case yields divergent behavior of the 2D effective interaction, akin to the unitary limit in three-dimensional atomic clouds. We derive an analytical expression for the interaction enhancement factor as a function of the polariton dipole moment and density, and we show that it can be very large in typical experimental conditions. Being exact in the dilute regime, this result ultimately holds for two polaritons in vacuum. Another interesting prediction of our theory is that the many-body polariton states

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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 condense into the paired state, which is the equilibrium ground state when the discrete level ε is below the scattering threshold. Second, the width of the resonance β must be sufficiently large for polaritons to feel the interior of the barrier during their lifetime τ. We shall assume a *broad* resonance, which seems to be the most likely for polaritons because of their very low effective mass [26]. Hence, we let

$$\beta \gg \varepsilon \tag{1}$$

and $\hbar/\beta \ll \tau \ll \tau_p$, where τ_p is the thermalization time [27].

The system is a mixture of two polariton flavors $\hat{c}_\sigma$ with $\sigma = (\uparrow, \downarrow)$ and their bipolaritonic pairs $\hat{C}$. In practice, "↑" and "↓" typically correspond to left- and right-circularly polarized photons [28]. In planar dielectric microcavities, the lower polariton dispersion is split into the linearly polarized transverse magnetic (TM) and transverse electric (TE) modes [29]. This splitting acts as an effective magnetic field that flips the polariton pseudospin σ. Provided that

$$p \ll \sqrt{m_{\mathrm{LT}}\beta}/\hbar, \tag{2}$$

the decay of a bound state due to the polariton spin-flip is subdominant with respect to the tunneling under the dipolar potential barrier. Here, the parameter m_{LT} accounts for both the bare photonic and low-k excitonic TE-TM splitting [30,31]. In confined geometries [32,33] one should let $p \sim \pi/L$, where L is the characteristic length of the confinement. Under the condition (2), we may consider a single polariton branch $E(\boldsymbol{p})$ characterized by the effective mass m in the vicinity of its minimum. Analysis of a possible departure from this approximation will be given in a separate paper.

The many-body Hamiltonian reads

$$\hat{H} = \sum_{\mathbf{p},\sigma} E(\boldsymbol{p})\hat{c}^\dagger_{\sigma,\mathbf{p}}\hat{c}_{\sigma,\mathbf{p}} + \sum_{\mathbf{k}}[2E(\boldsymbol{k}/2)+\varepsilon]\hat{C}^\dagger_{\mathbf{k}}\hat{C}_{\mathbf{k}} + \frac{g}{2S}\sum_{\mathbf{p}_1,\mathbf{p}_2,\mathbf{q},\sigma}\hat{c}^\dagger_{\sigma,\mathbf{p}_1+\mathbf{q}}\hat{c}^\dagger_{\sigma,\mathbf{p}_2-\mathbf{q}}\hat{c}_{\sigma,\mathbf{p}_1}\hat{c}_{\sigma,\mathbf{p}_2}$$
$$+\sqrt{\frac{\hbar^2\beta}{2\pi mS}}\sum_{\boldsymbol{k},\boldsymbol{p}}\left(\hat{c}^\dagger_{\uparrow,\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}\hat{c}^\dagger_{\downarrow,-\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}\hat{C}_{\boldsymbol{k}} + \hat{c}_{\uparrow,-\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}\hat{c}_{\downarrow,\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}\hat{C}^\dagger_{\boldsymbol{k}}\right). \tag{3}$$

Here the first two terms describe the dispersions of single polaritons and bipolaritons, respectively, with $\boldsymbol{p} = (p_x, p_y)$ and $E(\boldsymbol{p}) = \hbar^2 p^2/2m$ at the bottom of the band ($\boldsymbol{p} \to \mathbf{0}$). In the limit of zero exciton dipole moment, the discrete level ε can be identified with the binding energy of a loosely bound bipolariton molecule [34]. The next term is the usual background interaction between the polaritons with alike spins (accounting both for the short-range part and the dipolar tail of the bare exciton potential) in the quantization area S [35]. The last term models 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 [15,36].

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

$$i\hbar\frac{d\hat{c}_{\sigma,\boldsymbol{p}}}{dt} = [E(\boldsymbol{p})+\mu_\sigma]\hat{c}_{\sigma,\boldsymbol{p}} + \sqrt{\frac{\hbar^2\beta}{2\pi mS}}\sum_{\boldsymbol{k}}\hat{c}^\dagger_{\sigma'\neq\sigma,\boldsymbol{k}}\hat{C}_{\boldsymbol{k}+\boldsymbol{p}}, \tag{4a}$$

$$i\hbar\frac{d\hat{C}_{\boldsymbol{k}}}{dt} = [2E(\boldsymbol{k}/2)+\varepsilon]\hat{C}_{\boldsymbol{k}} + \sqrt{\frac{\hbar^2\beta}{2\pi mS}}\sum_{\boldsymbol{p}}\hat{c}_{\uparrow,-\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}\hat{c}_{\downarrow,\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}, \tag{4b}$$

where we have replaced the Hartree groups of operators by c-numbers and defined

$$\mu_\sigma = \frac{g}{S}\sum_{\boldsymbol{q}}|c_{\sigma,\mathbf{q}}|^2 = gn_\sigma, \tag{5}$$

with n_σ being the polariton densities in each component. By introducing the slowly varying amplitudes

$$\hat{c}_{\sigma,\boldsymbol{p}} = \hat{\mathfrak{c}}_{\sigma,\boldsymbol{p}}e^{-i[E(\boldsymbol{p})+\mu_\sigma]t/\hbar}, \tag{6}$$

we notice the existence of a stationary ($d\hat{\mathfrak{c}}_{\sigma,\boldsymbol{p}}/dt = 0$) solution of Eq. (4b) in the form

$$\hat{C}_{\boldsymbol{k}} = \frac{\sqrt{\frac{\hbar^2\beta}{2\pi mS}}}{\mu_\uparrow+\mu_\downarrow-\varepsilon'_{\boldsymbol{k}}}\sum_{\boldsymbol{p}}\hat{c}_{\uparrow,-\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}\hat{c}_{\downarrow,\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}, \tag{7}$$

where the motion of the bipolariton mode is reduced to that of a pair of polaritons with opposite spins. Here $\varepsilon'_{\boldsymbol{k}} = \varepsilon - \varepsilon_{\boldsymbol{k}}$ with

$$\varepsilon_{\boldsymbol{k}} = \frac{\sum_{\boldsymbol{p}}E_{\mathrm{rel}}(\boldsymbol{p},\boldsymbol{k})\langle\hat{\mathfrak{c}}_{\uparrow,-\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}\hat{\mathfrak{c}}_{\downarrow,\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}\rangle}{\sum_{\boldsymbol{p}}\langle\hat{\mathfrak{c}}_{\uparrow,-\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}\hat{\mathfrak{c}}_{\downarrow,\boldsymbol{p}+\frac{\boldsymbol{k}}{2}}\rangle} \tag{8}$$

being the kinetic energy of the relative motion in the pair, $E_{\mathrm{rel}}(\boldsymbol{p},\boldsymbol{k}) = E(\boldsymbol{p}+\boldsymbol{k}/2)+E(-\boldsymbol{p}+\boldsymbol{k}/2)-2E(\boldsymbol{k}/2)$.

The condition (1) provides a physical meaning to the solution (7). The objects $\hat{C}_{\boldsymbol{k}}$ should be regarded as auxiliary fields describing the onset of pair correlations between the polaritons, rather than new (quasi)particles. Indeed, substituting (7) into the last term of the Hamiltonian (3), one obtains an effective model

$$\hat{H}' = \sum_{\mathbf{p},\sigma}E(\boldsymbol{p})\hat{c}^\dagger_{\sigma,\mathbf{p}}\hat{c}_{\sigma,\mathbf{p}} + \frac{1}{2S}\sum_{\mathbf{p}_1,\mathbf{p}_2,\mathbf{q},\sigma,\sigma'}\hat{c}^\dagger_{\sigma,\mathbf{p}_1+\mathbf{q}}\hat{c}^\dagger_{\sigma',\mathbf{p}_2-\mathbf{q}}g_{\sigma\sigma'}\hat{c}_{\sigma,\mathbf{p}_1}\hat{c}_{\sigma',\mathbf{p}_2}, \tag{9}$$

with $g_{\uparrow\uparrow} = g_{\downarrow\downarrow} = g$ and

$$g_{\uparrow\downarrow} = \frac{\hbar^2}{2\pi m}\frac{\beta}{\left(\mu_\uparrow + \mu_\downarrow - \varepsilon'_{\boldsymbol{p}_1+\boldsymbol{p}_2}\right)}. \tag{10}$$

One can see that the pair correlations between the polaritons manifest themselves as resonantly strong two-body interactions. For a broad resonance considered in this work, the large magnitude of $g_{\uparrow\downarrow}$ is primarily due to the relation (1) (the μ_σ's are on the order of ε). However, by reducing the difference $\mu_\uparrow + \mu_\downarrow - \varepsilon'_{\boldsymbol{p}_1+\boldsymbol{p}_2}$ (by, e.g., decreasing the polariton density), one may also observe the typical resonant growth of the interaction strength. Though here we have in mind the case $\mu_\uparrow + \mu_\downarrow > \varepsilon'_{\boldsymbol{p}_1+\boldsymbol{p}_2}$, the formula (10) can be used on the attractive side $\mu_\uparrow + \mu_\downarrow < \varepsilon'_{\boldsymbol{p}_1+\boldsymbol{p}_2}$ as well.

Assume now a resonant excitation of a coherent mixture of "↑" and "↓" polaritons at some point $\boldsymbol{p}_0$ on the dispersion curve $E(\boldsymbol{p})$. The output signal is registered at the distance $l \sim v_{\rm gr}\tau$ from the excitation spot. Here $v_{\rm gr}$ is the corresponding group velocity. For simplicity, assume equal densities $n_\uparrow = n_\downarrow \equiv n = N/S$, which in practice may be achieved by using linearly polarized light. We therefore take the following initial ($t = 0$) configuration:

$$\hat{c}_{\sigma,p_0}(0)|\psi\rangle = \sqrt{N}\,|\psi\rangle\,, \quad \hat{c}_{\sigma,p\neq p_0}(0)\,|\psi\rangle = 0. \tag{11}$$

The bipolariton part of the many-body wave function $|\psi\rangle$ is initially in the vacuum state:

$$\hat{C}_{2p_0}(0)\,|\psi\rangle = 0. \tag{12}$$

By substituting the slowly varying c-numbers [see the definition (6)] $\mathsf{c}_{\sigma,p_0} = \rho_\sigma e^{i\phi_\sigma}$ and $\mathsf{C}_{2p_0} = \rho e^{i\phi}$ into Eqs. (4), and omitting the terms scaling as $\sqrt{\varepsilon/\beta}$, one can find

$$\phi(t) = \phi_\uparrow(t) + \phi_\downarrow(t) \pm \pi/2, \quad \phi_\sigma(t) = \phi_\sigma(0), \tag{13}$$

and

$$\rho_\sigma(t) = \sqrt{N}\cosh^{-1}(t/\tau_0), \quad \rho(t) = \sqrt{N}\tanh(t/\tau_0). \tag{14}$$

Equation (14) shows that on the characteristic timescale

$$\tau_0 = \sqrt{\frac{2\pi m}{\beta n}}, \tag{15}$$

the coherent mixture is entirely converted into the paired state (7). According to (9) and (10), the modified polariton blueshift is given by

$$\mu'_\sigma = gn + \frac{\hbar^2 n}{2\pi m}\frac{\beta}{(2ng - \varepsilon)}. \tag{16}$$

Note that spreading of the polariton distribution over the $\boldsymbol{k}$-space region where the dispersion can be approximated by a linear function will not affect the result (16) since, according to (8), in this region one has $\varepsilon_{\boldsymbol{k}} \equiv 0$.

The relation (1) refers to the typical case $d \sim d_c$, where d_c is the critical value at which the true bound state disappears [39,40]. In general, the width of the resonance changes from 0 to ∞ (the latter describing the ultimate case where the level washes out) as the exciton dipole moment is tuned from $d \ll d_c$ to $d \gg d_c$. For $0 \leqslant d \lesssim d_c$ one may take $\beta(d) = \mathcal{B}d$. The corresponding dependence for the position of the level has the form [15] $\varepsilon(d) = \mathcal{E}(d - d_c)$.

Following Ref. [22], one may then consider the "interaction enhancement factor" $\eta(d,n) = \mu'_\sigma/\mu_\sigma - 1$ as a function of the dipole moment d and density n. Substituting the above relations for $\beta(d)$ and $\varepsilon(d)$ into Eq. (16), we obtain

$$\eta(d,n) = \frac{\hbar^2}{2\pi mg}\frac{\mathcal{B}d}{[2ng - \mathcal{E}(d - d_c)]}. \tag{17}$$

Though our analytical methods do not allow us to estimate the typical values of the parameters $\mathcal{B}$ and $\mathcal{E}$, by virtue of (1) one may expect $\mathcal{B} \gg \mathcal{E}$ and, consequently, $\eta \gg 1$. The relation (1), in turn, is guaranteed by very low values of the polariton mass m as compared to bare excitons [26].

Interestingly, the strong correlations in the paired state (7) squeeze the polariton wave functions. To illustrate this point, consider again the situation discussed above, where one starts from a coherent state (11) for polaritons and a vacuum state (12) for their pairs. Introduce rotated quadratures

$$\begin{aligned}\hat{x}_\sigma &= \tfrac{1}{2}\left(\hat{\mathsf{c}}_{\sigma,p_0}e^{-i\phi_\sigma} + \hat{\mathsf{c}}^\dagger_{\sigma,p_0}e^{i\phi_\sigma}\right),\\ \hat{y}_\sigma &= \tfrac{1}{2i}\left(\hat{\mathsf{c}}_{\sigma,p_0}e^{-i\phi_\sigma} - \hat{\mathsf{c}}^\dagger_{\sigma,p_0}e^{i\phi_\sigma}\right),\end{aligned} \tag{18}$$

and

$$\begin{aligned}\hat{X} &= \tfrac{1}{2}(\hat{\mathsf{C}}_{2p_0}e^{-i\phi} + \hat{\mathsf{C}}^\dagger_{2p_0}e^{i\phi}),\\ \hat{Y} &= \tfrac{1}{2i}(\hat{\mathsf{C}}_{2p_0}e^{-i\phi} - \hat{\mathsf{C}}^\dagger_{2p_0}e^{i\phi}).\end{aligned} \tag{19}$$

Write $\hat{x}_\sigma = x_\sigma + \delta\hat{x}_\sigma$ and the same for $\hat{y}_\sigma$, $\hat{X}$, and $\hat{Y}$. The linearized equations of motion for the quadrature fluctuations read

$$\begin{aligned}\frac{d}{dt}\delta\hat{x}_{\uparrow,\downarrow} &= \pm\sqrt{\frac{\beta}{2\pi mS}}(\rho_{\downarrow,\uparrow}\delta\hat{X} + \rho\delta\hat{x}_{\downarrow,\uparrow}),\\ \frac{d}{dt}\delta\hat{y}_{\uparrow,\downarrow} &= \pm\sqrt{\frac{\beta}{2\pi mS}}(\rho_{\downarrow,\uparrow}\delta\hat{Y} - \rho\delta\hat{y}_{\downarrow,\uparrow}),\\ \frac{d}{dt}\delta\hat{X} &= \mp\sqrt{\frac{\beta}{2\pi mS}}(\rho_\uparrow\delta\hat{x}_\downarrow + \rho_\downarrow\delta\hat{x}_\uparrow),\\ \frac{d}{dt}\delta\hat{Y} &= \mp\sqrt{\frac{\beta}{2\pi mS}}(\rho_\uparrow\delta\hat{y}_\downarrow + \rho_\downarrow\delta\hat{y}_\uparrow),\end{aligned} \tag{20}$$

where the sign "+" or "−" corresponds to the two possible choices of the phase shift in Eq. (13), and ρ, ρ_σ are given by (14). At $t = 0$ one can use Eqs. (18) and (11) to find $\langle\delta\hat{x}^2_\sigma(0)\rangle = 1/4$ and $\langle\delta\hat{y}^2_\sigma(0)\rangle = 1/4$, the well-known property of a coherent state [41]. In contrast, at $\tau_0 \ll t \lesssim \tau$, where τ_0 is given by Eq. (15), one can substitute $\rho_\sigma = 0$ and $\rho = \sqrt{N}$ into the first pair of Eqs. (20) to obtain

$$\begin{aligned}\left\langle\delta\hat{x}^2_\sigma(t)\right\rangle &\sim e^{\pm t/\tau_0},\\ \left\langle\delta\hat{y}^2_\sigma(t)\right\rangle &\sim e^{\mp t/\tau_0},\end{aligned} \tag{21}$$

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

The requirement $\tau_0 \ll \tau$ sets the lower value of the polariton density at which the predicted effects may be observed. For sufficiently large β, one may operate in the ultradilute regime and even in the few-particle limit. In Eqs. (10), (16), and (17), the latter is formally achieved by letting $n = 0$. Strong repulsive interactions between just two polaritons may

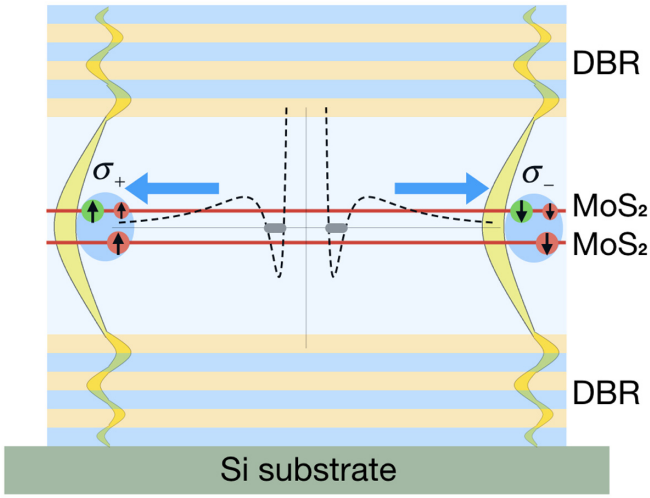

FIG. 1. Sketch of a TMD-based planar microcavity featuring unitary dipolar polaritons. The active region consists of a homobilayer structure with AA' or AB stacking, which allows dipolar excitons with strong oscillator strengths and convenient selection rules [37,38]. Black arrows show the spin orientation of the conduction and valence electronic states in the $K^{\pm}$ valleys. The holes are delocalised over both layers [37]. The "↑" and "↓" excitons couple to the cavity modes with opposite helicities. The dashed line traces the profile of the two-body interaction potential $V_{\uparrow\downarrow}(r)$ felt by each exciton [26]. This supports a quasidiscrete level (gray bar) separated from the outer continuum by a barrier due to the dipolar repulsion. The resulting polaritons experience strong repulsion and squeezing of their collective states.

be particularly promising for implementation of the dual-rail polariton logic [42].

The condition $\hbar/\beta \ll \tau$ 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 result in a loss of correlations and, at 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. [43]), which restores the significance of the result (21).

Our last remark concerns the choice of the sign in Eq. (13). Under the condition (1), the Josephson coupling of the polariton states to the resonance 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 (21) experimentally.

In addition to the already mentioned state-of-the-art in QW microcavities [21–23], in Fig. 1 we sketch a possible implementation of unitary polaritons with TMDs. A convenient choice may be a homobilayer of MoX_2, showing large oscillator strengths and spin-valley selection rules analogous to the monolayer excitons [37,38]. Natural separation between the layers here is close to the threshold d_c, the latter being on the order of the exciton Bohr radius [15]. Tuning of the resonance position in this case of fixed d may be achieved by varying the magnitude of the satellite carrier wave function in one of the layers (the residue of the intralayer exciton) [37].

To conclude, we predict anomalously large enhancement of repulsive interactions in a system of dipolar polaritons. 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, such as atomically thin layers of TMDs and quantum wells. 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 [44]. The idea of using the shape resonance to produce strong pair correlations and squeezing at ultralow polariton densities opens wide perspectives for future research and applications. Thus, an interesting direction would be application of the physics discussed in this work to the recently established field of topological polaritons [45,46].

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