Polariton-polariton interaction beyond the Born approximation: A toy model study

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(Dated: April 14, 2020)

We theoretically investigate the polariton-polariton interaction in microcavities beyond the commonly used Born approximation (i.e., mean-field), by adopting a toy model with a contact interaction to approximately describe the attraction between electrons and holes in quantum well and by using a Gaussian pair fluctuation theory beyond mean-field. We obtain a density or chemical potential independent polariton-polariton interaction strength due to the populated photon field, in contrast to the picture of a weakly interacting two-dimensional Bose gas of exciton-polaritons. We show that the beyond-mean-field or beyond-Born-approximation effect typically leads to a factor of about 2 reduction in the polariton-polariton interaction strength. Together with the saturation effect at very strong light-matter coupling, we find that the standard expression \( g_{PP}^{(0)} = X^2 g_{XX}^{(0)} \), where \( X^2 \) is the excitonic Hopfield coefficient in the exciton-polariton model and \( g_{XX}^{(0)} \) is the exciton-exciton interaction strength within the Born approximation, typically overestimates the polariton-polariton interaction strength by a factor of about 3 under current experimental conditions. We compare our prediction with the most recent measurement and argue that the beyond-Born-approximation effect to the polariton-polariton interaction strength is crucial for a quantitative understanding of the experimental data by E. Estrecho et al., Phys. Rev. B 100, 035306 (2019).

I. INTRODUCTION

Exciton-polaritons in microcavities are half-light and half-matter bosonic quasiparticles \([1,2]\), arising from the strong coupling between the photon field and tightly-bound electron-hole pairs (i.e., excitons). Due to the ultra-small effective mass inherent from the light, a Bose-Einstein condensate (BEC) of exciton-polaritons can occur at extremely high temperature and therefore may provide an ideal platform to realize new technologies such as optical switches and optical transistors, which should radically change the future optical communications \([3,4]\). To this aim, it is crucial to determine and manipulate the polariton-polariton interaction and to reach the strongly interacting regime for photons \([5,6]\). Unfortunately, so far the experimental measurements and theoretical understandings of the polariton-polariton interaction in microcavities are not satisfactory, despite enormous efforts over the past two decades.

The difficulty in the experimental determination of the polariton-polariton interaction strength comes from the strong influence by the reservoir of high-energy excitons created and maintained by the optical pump, which is necessary to seed polaritons to form a BEC \([1]\). The residual polariton-reservoir interaction together with the quantum confinement effect due the optical pump leads to a large uncertainty in determining the polariton-polariton interaction strength \([10]\). This is clearly seen from the previous values obtained by various techniques such as the ground-state blue shift \([10-14]\) and the first- and second-order correlation functions \( g^{(1)}(\tau) \) and \( g^{(2)}(\tau) \) \([8,9,15]\), which scatter over a range spanning almost four orders in magnitude (see, i.e., Fig. 5(b) in Ref. \([10]\)). In an extreme case \([13]\), the measured polariton-polariton interaction strength can be about two orders of magnitude larger than the theoretical predictions \([16-18]\), making theoretical understanding very challenging. This bottleneck in measurement has recently been overcome by Estrecho and his collaborators \([10]\). The spatial overlap between the polariton condensate and the reservoir is carefully removed and the condensate is put into a single-mode Thomas-Fermi regime under a box potential. By taking a linear dependence of the blue shift of the ground-state energy \( \Delta E_0 \) on the peak particle density \( n_{TF} \),

\[
\Delta E_0 = g_{PP} n_{TF},
\]

the polariton-polariton interaction strength \( g_{PP} \) (i.e., the slope of the blue shift) is then determined as a function of the photon detuning \([10]\). Although there is still a notable error bar in measured \( g_{PP} \) due to the systematic error in the density calibration, it is arguably the most accurate measurement so far. Hence, we may treat this measurement as a benchmark result.

On the theoretical side, there are also several difficulties in understanding and calculating the polariton-polariton interaction strength. First, strictly speaking the linear behavior between the ground-state energy shift and the particle density does not hold, if we treat a system of exciton-polaritons confined in microcavities as an interacting two-dimensional (2D) Bose gas \([20]\). According to the Bogoliubov theory, the relation between the chemical potential \( \mu_B \) and the number density \( n \) of an interacting 2D Bose gas would be given by \([21]\),

\[
n \simeq \frac{m_B \mu_B}{4 \pi \hbar^2} \ln \left[ \frac{4 \hbar^2}{m_B \mu_B a_s^2 e^{2 \gamma + 1}} \right],
\]

where \( m_B \) is the mass of bosons, \( a_s \) is the 2D s-wave scattering length for the short-range (contact) interaction between bosons, and \( \gamma \approx 0.577 \) is Euler’s constant.
This indicates a density or chemical potential dependent interaction strength
\[
g(\mu_B) = \frac{\mu_B}{n} = \frac{4\pi\hbar^2}{m_B} \ln^{-1} \left[ \frac{4\hbar^2}{m_B \mu_B a_X^2 e^{2\gamma+1}} \right]. \tag{3}
\]
In particular, towards the dilute limit the interaction strength would vanish due to the vanishingly small chemical potential and density. This result apparently disagrees with the linear dependence observed or assumed in experiments. Secondly, to date most calculations of the exciton-exciton interaction strength are based on the Born approximation, which gives a constant, density independent interaction strength. In the exciton-polariton model, it takes the form [17],
\[
g_{PP}^{(0)} = X_{LP}^4 g_{XX}^{(0)}, \tag{4}
\]
where
\[
g_{XX}^{(0)} \simeq 6.06E_X a_X^2 = 6.06\hbar^2/M \tag{5}
\]
is the exciton-exciton interaction strength in 2D and
\[
X_{LP}^2 = \frac{1}{2} \left[ 1 + \frac{\delta/2}{\sqrt{\delta^2/4 + \Omega^2}} \right] \tag{6}
\]
is the excitonic Hopfield coefficient with the photon detuning \( \delta \) (measured with respect to the exciton energy \( -E_X \)) and the light-matter coupling \( \Omega \). Here, \( a_X \) and \( E_X = \hbar^2/(Ma_X^2) \) are the Bohr radius and binding energy of excitons, respectively, and we have assumed for simplicity that electrons and holes take the same mass \( m_e = m_h = m_{eh} = M \). We have also used the superscript "0" to explicitly indicate the results within the Born approximation. Eq. (4) is very easy to understand since the interaction between polaritons is mediated by the excitonic component of polaritons only. However, it should be corrected when the light-matter coupling \( \Omega \) becomes strong and comparable to \( E_X \), so that the standard exciton-polariton model starts to break down. This so-called oscillator strength saturation effect is well-known in the literature [12, 17] and most recently has been rigorously treated by solving the exact two-body problem of the underlying fermionic electron-hole-photon Hamiltonian in the dilute limit [22]. It was shown that the correction to Eq. (4) can be about \( \sim 20\% \) at the very strong coupling regime when \( \Omega \sim E_X \). On the other hand, experimentally, the exciton-exciton interaction strength \( g_{XX}^{(0)} \) in Eq. (4) may also need revision, considering the quasi-2D configuration of the quantum well, whose width \( l_z \) would be similar to \( a_X \) [10]. In such a situation, a rough estimation gives rise to,
\[
g_{XX, a_{2D}}^{(0)} = \frac{26\pi}{3} E_X a_X^2 \left( \frac{a_X}{l_z} \right). \tag{7}
\]
This expression was used by Estrecho and his collaborators to set a theoretical upper bound for the polariton-polariton interaction strength [10]. It is about three times larger than the measured value.

It is certainly not satisfactory to restrict theoretical analysis just to the Born approximation. This is particularly relevant in 2D, where quantum and thermal fluctuations are so significant that the equation of state of the system can qualitatively be altered [22]. The density or chemical potential interaction strength of an interacting 2D Bose gas mentioned in the above is already an excellent example. Even in three dimensions (3D), the beyond-Born-approximation effect could be very significant. A well-known case is a two-component ultracold atomic Fermi gas with a contact interaction characterized by a 3D s-wave length length \( a_F \). In the Born limit where tightly bound molecules are formed, the exact molecule-molecule scattering length is \( a_s \sim 0.66a_F \), much smaller than the result \( a_s^{(0)} = 2a_F \) obtained within the Born approximation.

In this work, we aim to better understand the polariton-polariton interaction in 2D by going beyond the Born approximation. This is possible if we replace the Coulomb interaction between electrons and holes with a short-range contact interaction, whose scattering length is tuned to correctly reproduce the binding energy of excitons. Therefore, we are able to construct a toy model for the electron-hole-photon system, which captures the important underlying fermionic degree of freedom of exciton-polaritons. By applying a Gaussian pair fluctuation theory (GPF) beyond mean-field as in the previous investigation [26, 27], much smaller than the result \( a_s^{(0)} = 2a_F \) obtained within the Born approximation.

In the rest of this work, we will discuss the two-component ultracold atomic Fermi gas, which is represented by the toy model [28]. In particular, towards the dilute limit the interaction strength would vanish due to the vanishingly small chemical potential and density. This result apparently disagrees with the linear dependence observed or assumed in experiments.
for the exciton-polariton system in microcavities. In Sec. III, we consider the case with a small light-matter coupling and a large photon detuning, for which a weakly interacting 2D exciton condensate is recovered. We discuss the exciton-exciton interaction within the Born approximation (i.e., mean-field level) and beyond the Born approximation (i.e., GPF level). In Sec. IV, we investigate the polariton system at large light-matter couplings and define a generalized excitonic Hopfield coefficient, which captures the oscillator strength saturation effect and the reduced size of exciton wave-functions due to the photon-mediated attraction [22]. We show that the correction to the polariton-polariton interaction strength beyond the Born approximation might be characterized by using the mean-field density fractions. In Sec. V, we assume the insensitivity of the beyond-Born-approximation effect on the underlying electron-hole-hole and compare our prediction with the latest measurement of the polariton-polariton interaction strength [14]. Finally, we summarize in Sec. VI.

II. THEORETICAL MODEL AND GAUSSIAN PAIR FLUCTUATION THEORY

The 2D electron-hole-photon system in microcavities can be described by the model Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{LM} + \mathcal{H}_C$ as [29 31]

$$\mathcal{H}_0 = \sum_{k\sigma} \epsilon_k c^\dagger_{k\sigma} c_{k\sigma} + \sum_{q} \left[ \frac{\hbar^2 q^2}{2m_{ph}} + \delta_0 - \mu \right] \phi^\dagger_q \phi_q, \quad (8)$$

$$\mathcal{H}_{LM} = \frac{g_0}{\sqrt{S}} \sum_{kq} \left[ \phi^\dagger_q c_{k\sigma}^\dagger c_{-k\sigma}^\dagger + h.c. \right], \quad (9)$$

$$\mathcal{H}_C = \frac{1}{2S} \sum_{kk'q} V_{kk'}^{\sigma\sigma'} c^\dagger_{k\sigma} c^\dagger_{-k\sigma} c_{-k'\sigma'} c_{k'\sigma'}, \quad (10)$$

where $\xi_k \equiv \hbar^2 k^2/(2M) - \mu/2$, $\delta_0$, $\mu$, $g_0$ and $S$ are the electronic dispersion within an effective mass approximation, bare cavity detuning, chemical potential, bare light-matter coupling strength, and the area of the system, respectively. We have taken the same mass $M = m_{eh} \simeq 0.067m_0$ for electrons and holes (where $m_0$ is the free-electron mass) and an ultra-small photonic mass $m_{ph} \simeq 3 \times 10^{-5}m_0$ due to the microcavity confinement [2]. $c_{k\sigma}$ are the annihilation operators of electrons ($\sigma = e$) and holes ($\sigma = h$), and $\phi_q$ denote the annihilation operators of photons.

In Eq. (10), $V_{kk'}^{\sigma\sigma'}$ are the Coulomb-like interactions among electrons and holes, and are defined as the Fourier transformation of a screened potential [32 33],

$$V_{\sigma\sigma'}^C(r) = \chi_{\sigma\sigma'} \frac{\pi e^2}{2\varepsilon_s r_{0}} \left[ H_0 \left( \frac{r}{r_{0}} \right) - Y_0 \left( \frac{r}{r_{0}} \right) \right], \quad (11)$$

where $\chi_{\sigma\sigma'} = \mp 1$ for $\sigma = \sigma'$ and $\chi_{\sigma\sigma'} = \mp 1$ for $\sigma \neq \sigma'$, $\varepsilon_s$ is the dielectric constant of the substrate surrounding the quantum well, $H_0(x)$ and $Y_0(x)$ are respectively the Struve and Neumann functions, and $r_0$ is an effective screening length. This particular form of the Coulomb-like interaction is due to the large difference in the dielectric constants of the quantum well and of the substrate, which strongly modifies the Coulomb interaction at short distance [32 33]. The model Hamiltonian is extremely difficult to solve because of the non-local nature of the Coulomb interaction. To find a way around, we propose a toy model by replacing the Coulomb interaction with a local contact interaction [31 33], i.e.,

$$\mathcal{H}_C = \frac{u_0}{S} \sum_{kk'q} c^\dagger_{k\sigma} c_{k'\sigma'} c^\dagger_{k'\sigma} c_{k\sigma} - k'h^2 c^\dagger_{k\sigma} + k'c_{k\sigma'}, \quad (12)$$

where the interaction strength $u_0$ should be tuned to reproduce the correct ground-state energy of excitons with the Coulomb-like interaction Eq. (11).

It is useful to note that, in ultracold atomic physics, our toy model Hamiltonian describes a two-component interacting Fermi gas near Feshbach resonances at the crossover from a BEC to a Bardeen–Cooper–Schrieffer (BCS) superfluid [35–38]. The Feshbach coupling is simply the light-matter coupling here. The photons now play the role of the closed-channel molecules, if we ignore a small modification to the photon mass (i.e., we cannot have the relation $m_{ph} = 2M$, which holds for ultracold atoms), while the excitons at low density correspond to the tightly-bound Cooper pairs in the open channel. At a broad Feshbach resonance, which is realized when the light-matter coupling is infinitely strong, the toy model Hamiltonian has actually been investigated experimentally [39 41] and theoretically [42 43]. Here, the purpose of this work is to understand the molecular scat-tering length in the case of a very strong yet finite light-matter coupling or Feshbach coupling, which is not explored so far in the context of ultracold atoms.

The use of contact interactions both for the electrons and holes ($u_0$) and for the light-matter coupling ($g_0$) will lead to an ultraviolet divergence. This divergence can be formally removed by the so-called regularization procedure, after which the bare parameters $u_0$, $g_0$, and $\delta_0$ will be replaced by $u$, $g$, and $\delta = E_{\text{cav}} - (-E_X)$, respectively. Here, $E_{\text{cav}}$ is the cavity energy, and the renormalized parameters $u$ and $g$ are explicitly related to the physical observables of the exciton binding energy $E_X$ and the Rabi coupling $\Omega$ as follows [26]:

$$u = \frac{4\pi \hbar^2}{M} \ln^{-1} \left( \frac{E_X}{\varepsilon_0} \right), \quad (13)$$

$$g = 2\sqrt{\pi} \Omega u X \ln^{-1} \left( \frac{E_X}{\varepsilon_0} \right), \quad (14)$$

where $\varepsilon_0 \ll E_X$ is an unimportant energy scale used to regularize the logarithmic infrared divergence commonly encountered in two dimensions. For more details on the renormalization, we refer to Supplemental Material of Ref. [26], which also explains the solution of the two-particle problem.
To obtain the polariton-polariton interaction strength (which is intrinsically a six-particle problem, involving two photons, two electrons and two holes), we solve our toy model Hamiltonian using the many-body GPF theory \[23, 44–46\] and then consider the low-density dilute limit. The details of the GPF formalism are again outlined in Ref. [26]. Here, for self-containedness we briefly review the main equations. Taking the Hubbard–Stratonovich transformation, we first introduce a pairing field to decouple \(H_c\) in Eq. (12) and integrate out the fermionic fields \(c_{\kappa}\). We then obtain an effective action for the pairing field and photon field, whose superposition could be understood as a polariton field. At zero temperature, the saddle-point solution of the polariton field gives rise to a mean-field thermodynamic potential \[26\],

\[
\Omega_{\text{MF}} = \frac{\Delta^2}{u_{\text{eff}}} + \sum_k \left[ \xi_k - E_k + \frac{\Delta^2}{\hbar^2 k^2/M + \varepsilon_0} \right],
\]

where \(\Delta\) is an order parameter satisfying the gap equation \(\partial \Omega_{\text{MF}} / \partial \Delta = 0\),

\[
u_{\text{eff}} = u + g^2/\mu - \delta
\]

is an effective interaction incorporating the photon-mediated attraction, and \(E_k = \sqrt{\xi_k + \Delta^2}\) is the dispersion relation for fermionic Bogoliubov quasi-particles.

To go beyond mean-field, we expand the effective action in the BEC limit, the system can be viewed as a weakly interacting Bose gas of molecules \[23, 39\], with mass \(m_B = 2M\) and density \(n = n_F/2\) (\(n_F\) is the density of fermions). The exact four-body calculation shows that the molecular scattering length \(a_s\) related to the 2D scattering length between fermions \(a_{2D}\) through \[48\]

\[
\kappa a_{2D} \simeq 0.56 a_{2D}.
\]

One advantage of our GPF theory is that it can provide a reliable equation of state at zero temperature \[23, 44–46\]. In particular, in the dilute limit it gives an approximate but reasonably accurate molecular scattering length. For example, for a two-component interacting Fermi gas at BEC-BCS crossover in three dimensions, the molecular scattering length predicted by the GPF theory is about \(a_s \simeq 0.55 a_F\) \[44–46\], which is slightly smaller than the exact value \(a_s \simeq 0.60 a_F\) \[24\]. In two dimensions of interest, the GPF theory also provides a very accurate molecular scattering length \[23\], as we shall discuss in detail in the next section.

### III. 2D EXCITON CONDENSATE

For an interacting 2D Fermi gas with a contact interaction in the BEC limit, the system can be viewed as a weakly interacting Bose gas of molecules \[23, 39\], with mass \(m_B = 2M\) and density \(n = n_F/2\) (\(n_F\) is the density of fermions). The exact four-body calculation shows that the molecular scattering length \(a_s\) related to the 2D scattering length between fermions \(a_{2D}\) through \[48\]

\[
\kappa a_{2D} \simeq 0.56 a_{2D}.
\]

Here, \(a_{2D} = 2e^{-\gamma} a_X\) can be calculated by using the binding energy \(E_X = 4\hbar^2/(M a_{2D}^2 e^2)\). The molecular scattering length determined from the 2D GPF theory coincides with the exact value if we keep the two significant digits \[23\]. According to the Bogoliubov theory of a 2D weakly interacting Bose gas, Eq. (2), we thus obtain,

\[
n_B \simeq \frac{1}{2\pi a_X^2} \left[ -2 \ln \kappa - (\ln 2 + 1) - \ln \frac{\mu_B}{E_X} \right] \frac{\mu_B}{E_X}.
\]

In contrast, the mean-field theory cannot predict qualitatively correct equation of state. By writing the molecular chemical potential \(\mu_B\) in terms of the chemical potential of fermions \(\mu_F = \mu/2\) (i.e., \(\mu_B = 2m_F + E_X = \mu + E_X\)), from the mean-field equation of state

\[
\mu_F + \frac{E_X}{2} = \varepsilon_F \equiv \frac{\hbar^2 (2\pi n_F)}{2M}.
\]
we find that,

$$\mu_B = \frac{4\pi \hbar^2 n}{M} = 4\pi E_X a_X^2 n, \quad (25)$$

implying a molecule-molecule interaction strength $g_m = 4\pi E_B a_B^2$ within the Born approximation.

In the case that the photon field is not occupied, our toy model describes exactly the 2D interacting Fermi gas and molecules discussed in the above can be viewed as excitons. Hence, we find that the exciton-exciton interaction strength in the toy model within the Born approximation is,

$$g_{XX}^{(0)} = 4\pi E_X a_X^2, \quad (26)$$

which is about two times the exciton-exciton interaction strength in Eq. (1) when a Coulomb interaction is considered. To go beyond the Born approximation, we consider the GPF calculation at a small light-matter coupling $\Omega = 0.2E_X$ and a large photon detuning $\delta = 8E_X$, so the photon field is essentially not populated and the system could be a perfect weakly interacting 2D BEC of excitons in the dilute limit.

In Fig. 1 we show the density equation of state for small total density $n = n_{\text{tot}}$ or small chemical potential $\mu_B = \mu - E_{LP}$, where $E_{LP} = -E_X$ is the energy of the lower-polariton branch in the dilute limit in the absence of the photon field. We find that the mean-field (empty squares) and GPF results (solid circles) are indeed accurately described by Eq. (25) and Eq. (26), respectively. We emphasize that, within the GPF theory, the chemical potential dependent exciton-exciton interaction strength is given by,

$$g_{XX} (\mu_B) = \frac{2\pi E_X a_X^2}{-2 \ln \kappa - (\ln 2 + 1) - \ln(\mu_B/E_X)}. \quad (27)$$

It vanishes logarithmically in the zero-density limit, i.e., $g_{XX} (\mu_B \to 0) = 0$.

**IV. 2D EXCITON-POLARITON CONDENSATE**

What happens if the photon field is significantly occupied? In Fig. 2 we show the mean-field and GPF density
equations of state at zero photon detuning \( \delta = 0 \) and
two light-matter couplings \( \Omega = 0.2E_X \) (a) and \( \Omega = 0.8E_X \) (b). For comparison, we show also the corresponding
equations of state predicted by the exciton-polariton model (see, i.e., Eq. (14)) using black dashed line and red solid line, respectively. There are two interesting observations. First, the mean-field result apparently deviates from the anticipated behavior \( g_{PP}^{(0)} = X_{LP}^{(0)}g_X^{(0)} \) [17], indicating the breakdown of the exciton-polariton model. This deviation becomes larger when we increase the light-matter coupling. On the other hand, the GPF result clearly shows a linear dependence of the density on the chemical potential, suggesting the existence of a constant polariton-polariton interaction strength. Therefore, beyond mean-field the polariton system cannot be simply viewed as a weakly interacting 2D Bose gas of polaritons.

A. Born approximation (mean-field)

Let us first analyze the mean-field results. From the mean-field thermodynamic potential Eq. (15), we may derive the gap equation,

\[
\sqrt{\mu^2 + 4\Delta^2} - \mu = 2\varepsilon_0 \exp \left( \frac{4\pi\hbar^2}{M_u} \right),
\]

and the number equation,

\[
n_{MF} = \left( \frac{g}{\delta - \mu} \right)^2 \frac{\Delta^2}{u^{efi}} + \frac{M}{8\pi\hbar^2} \left( \sqrt{\mu^2 + 4\Delta^2} + \mu \right).
\]

In the dilute BEC limit, both the bosonic chemical potential \( \mu_B = \mu - E_{LP} \) and the order parameter \( \Delta \) are small controllable parameters, compared with the low-polariton energy \( E_{LP} \sim -E_X \). To the leading order, we thus have

\[
u_{efi} \to u + \frac{g^2}{E_{LP} - \delta} \equiv u_{LP}.
\]

Taylor-expanding the gap equation, we find that

\[-E_{LP} - \mu_B - \frac{\Delta^2}{E_{LP}} = \varepsilon_0 \exp \left( \frac{4\pi\hbar^2}{M_u} \right) \left[ 1 + \frac{\mu_B}{A} \right],
\]

where

\[A^{-1} \equiv \frac{4\pi\hbar^2}{M_u^2} \frac{g^2}{(\delta - E_{LP})^2}.
\]

The leading term of the above gap equation is simply the expression for the lower-polariton energy \( E_{LP} \), i.e., \( E_{LP} = -\varepsilon_0 e^{4\pi\hbar^2/(M_u)} \). Using this to eliminate the cut-off energy scale \( \varepsilon_0 \), we obtain

\[-\frac{\Delta^2}{E_{LP}} = \left[ 1 - \frac{E_{LP}}{A} \right] \mu_B.
\]

Next, to the leading order the number equation can be casted into the form,

\[n_{MF} = \frac{M}{4\pi\hbar^2} \left[ 1 - E_{LP} - \frac{\Delta^2}{E_{LP}} \right].
\]

By using the fact that \( n = n_{tot} = n_{MF} \) within mean-field and by combining these two equations to remove the pairing gap \(-\Delta^2/E_{LP}\), we find that,

\[\mu_B = \left[ 1 - \frac{E_{LP}}{A} \right]^{-2} \left( 4\pi E_X a_X^2 \right) n.
\]

It is readily seen that, the polariton-polariton interaction strength within the mean-field (Born approximation) is given by,

\[g_{PP}^{(0)} = \xi_{LP}^2 \left( 4\pi E_X a_X^2 \right),
\]

where we have defined,

\[\xi_{LP}^2 \equiv \left[ 1 - \frac{E_{LP}}{A} \right]^{-1} \left[ 1 - \left( 4\pi^2 \hbar^2 \right) g_{X} \frac{2E_{LP}}{M^2 u_{LP}^2 (\delta - E_{LP})^2} \right]^{-1}.
\]

By recalling that \( 4\pi E_X a_X^2 = g_X^{(0)} \) is the exciton-exciton interaction strength for our toy model, we may interpret \( \xi_{LP}^2 \) as a generalized exciton Hopfield coefficient. This interpretation can be easily examined for a small light-matter coupling, at which the exciton-polariton model is applicable. For a small Rabi coupling \( \Omega \ll E_X \), we may approximate \( u_{LP} \simeq u \) and use the expression for the lower polariton energy,

\[E_{LP} = \frac{\delta}{2} - \sqrt{\frac{\delta^2}{4} + \Omega^2}.
\]

By further taking \( g^2/u^2 = M\Omega^2/(4\pi\hbar^2 E_X) \), we find that,

\[\xi_{LP}^2 \simeq \left[ 1 + \left( \frac{\Omega^2}{(\delta - E_{LP})^2} \right)^{-1} \right] = X_{LP}^2.
\]

Thus, in the case of a small light-matter coupling, \( \xi_{LP}^2 \) reduces to \( X_{LP}^2 \), as we anticipate. An alternative explanation for the generalized exciton Hopfield coefficient \( \xi_{LP}^2 \) is given in Appendix A, where we consider the electron-hole vertex function or the polariton Green function.

In Fig. 3, we report the Hopfield coefficients \( X_{LP}^2 \) (black dashed line) and \( \xi_{LP}^2 \) (red solid line) as a function of the photon detuning at three light-matter couplings: \( \Omega = 0.1E_X \) (a), \( \Omega = 0.5E_X \) (b), and \( \Omega = 1.0E_X \) (c). At small coupling \( \Omega \ll E_X \) as shown in (a), \( \xi_{LP}^2 \) is essentially the same as the \( X_{LP}^2 \), as we have already confirmed analytically. However, as the light-matter coupling increases, \( \xi_{LP}^2 \) becomes increasingly smaller than \( X_{LP}^2 \) and the relative reduction can be about a few 10% when the light-matter coupling is comparable to the exciton binding energy \( \Omega \sim E_X \).
The difference between $\xi_{LP}^2$ and $X_{LP}^2$ at nonzero light-matter coupling is expected. For the Coulomb interaction $V_C(r) \propto -1/r$, it was understood in most previous works as the oscillator strength saturation effect and its explicit form at the order of $\Omega/E_X$ was derived analytically. The saturation correction enhances the polariton-polariton interaction strength. This difference was also numerically investigated by Levinsen and coworkers most recently. In addition to the known saturation correction, a more dramatic effect of light-matter coupling was revealed. At large light-matter coupling, the photon-mediated attraction becomes dominant between electrons and holes. As a result, the size of excitons in the low-polariton branch shrinks considerably and the exchange processes (for electrons or holes between two different polaritons, which is responsible for polariton-polariton repulsion) becomes less efficient. For our toy model with a contact interaction between electrons and holes, the reduction in the exchange processes seems to overwhelm the enhancement due to the saturation in the oscillator strength, leading to an overall smaller $\xi_{LP}^2$ in comparison with $X_{LP}^2$.

B. Beyond the Born approximation (GPF)

Here, we turn to consider the beyond-Born-approximation effect using the GPF theory. Naively, we argue that the polariton system consists of different types of carriers, as characterized by $n_{MF}$ and $n_{GPF}$, which are contributed from the mean-field saddle point and from pair fluctuations around the saddle point, respectively. In the case of completely suppressed fermionic degree of freedom, i.e., $n_{MF} \ll n_{GPF}$, the system could be viewed as a weakly-interacting Bose gas of exciton-polaritons, as we have already discussed in Sec. III. This picture is not true for the general case when the photon field starts to get occupied. In general, as shown in Appendix B, we find that both $n_{MF}$ and $n_{GPF}$ become significant and towards the zero-density limit, their ratio $n_{MF}/n_{GPF}$ saturates to a constant. At large light-matter coupling and near zero photon detuning, therefore, we may define a quantity,

$$\mathcal{F}_{BB} = \lim_{\mu_B \to 0} \left( \frac{n_{MF}}{n_{tot}} \right),$$

which itself is functions of the light-matter coupling $\Omega$ and of the photon detuning $\delta$. Now, using Eq. (34) and Eq. (33) for $n_{MF}$, in the zero-density limit we find,

$$\mu_B = 4\pi E_X a_X^2 \xi_{LP}^4 n_{MF} = \xi_{LP}^4 \mathcal{F}_{BB} (4\pi E_X a_X^2) n,$$

which implies a polariton-polariton interaction strength,

$$g_{PP} = (\xi_{LP}^4 \mathcal{F}_{BB}) \left( 4\pi E_X a_X^2 \right).$$

In other words, within GPF the polariton-polariton interaction strength is reduced by a factor of $\mathcal{F}_{BB}^{-1}$, compared with the Born approximation result $g_{PP}^{(0)} = \xi_{LP}^4 (4\pi E_X a_X^2)$. The linear dependence of the GPF result, as shown in Fig. 2(b), means that the mean-field contribution (i.e., the fermionic degree of freedom and condensed photons) is significant. Otherwise, the reduction factor $\mathcal{F}_{BB}$ will go to zero and the polariton-polariton interaction strength $g_{PP}$ becomes zero. The polariton system then crosses smoothly over to a weakly interacting 2D Bose gas of exciton-polaritons, as we discuss in Sec. III.

C. Comparison to the numerical results

We can now understand the two observations made at the beginning of this section, by using the main result of

FIG. 3. Exciton Hopfield coefficient $X_{LP}^2$ (black dashed line) and the generalized exciton Hopfield coefficient $\xi_{LP}^2$ (red solid line) as a function of the photon detuning $\delta$ at three light-matter couplings: $\Omega = 0.1E_X$ (a), $0.5E_X$ (b), and $1.0E_X$ (c).
function of the number density (in units of \( \text{\textxi} \)) where \( \text{\textxi} \) is responsible for the large light-matter coupling and \( \mathcal{F}_{\text{BB}} \) accounts for the beyond-Born-approximation effect. In Fig. 4, we replot Fig. 2(b) and add the anticipated behavior Eq. (42) for the mean-field result (black dashed line) and Eq. (43) for the GPF result (red solid line). It is clear that in the low-density limit, our analytic equations provide a satisfactory explanation to the numerical results, obtained using either mean-field or GPF theories.

D. Comparison to the analytic Bogoliubov result at small light-matter coupling

At small light-matter coupling, where the exciton-polariton model is applicable, the polariton-polariton interaction strength can be analytically obtained by using the Bogoliubov theory. Taking the equal mass for electrons and holes and the known exciton-exciton s-wave scattering length \( a_s = 2\kappa e^{-\gamma a_X} \) (where \( \gamma \approx 0.56 \)) as discussed in Sec. III for a contact electron-hole attraction, it takes the form of Eq. (43),

\[
\frac{g_{PP}}{g_{XX}^{(0)}} = \frac{\xi_{LP}^2}{2 \ln |E_X / (-E_{LP})| - 4 \ln (2\kappa)} \]

where \( E_{LP} \) is given by Eq. (38). At small light-matter coupling, we have \( \xi_{LP}^2 = X_{LP}^2 \). Therefore, by comparing this work,

\[
\frac{g_{PP}}{g_{XX}^{(0)}} = \frac{\xi_{LP}^2 \mathcal{F}_{\text{BB}}}{4 \ln (2\kappa)} \]

Eq. (43) and Eq. (44), we obtain that for \( \Omega \ll E_X \),

\[
\mathcal{F}_{\text{BB}} = \frac{1}{2 \ln |E_X / (-E_{LP})| - 4 \ln (2\kappa)} \]

In Fig. 5 we compare the numerical GPF result and the analytic Bogoliubov prediction for the polariton-polariton interaction strength (measured in units of \( g_{XX}^{(0)} \)) as a function of the photon detuning at \( \Omega = 0.1E_X \). A good agreement is found. Although two different theories with entirely different model Hamiltonians (i.e., fermionic vs. bosonic) are used, both of them reliably describe the exciton-polariton physics at small light-matter coupling.

V. COMPARISON TO THE EXPERIMENT

Although our main result Eq. (43) is obtained by using a toy model Hamiltonian with a contact interaction for electrons and holes, it would be interesting to see its relevance to the experimental measurements, where a Coulomb-like interaction, i.e., Eq. (11), should be considered. To this aim, let us make a bold assumption that, Eq. (43) depends very weakly on the underlying interaction between electrons and holes.

How can we assume that the beyond-Born-approximation effect should lead to the same reduction factor in the polariton-polariton interaction strength, for both contact interaction and Coulomb interaction? This is certainly difficult to justify. But, we may consider the exciton-exciton interaction strength in 3D, which seems to be the only example available for checking at the moment. According to a recent fixed-node diffusion Monte Carlo simulation with Coulomb interaction in 3D \( \gamma \), the exciton-exciton scattering length is about \( a_s = 1.5a_X \). Here, for a single exciton, its ground state
energy $E = -\hbar^2/(Ma_X^2)$. The Born approximation result for the exciton-exciton scattering length can be extracted from the expression,

$$g_{XX}^{(0)} = \frac{26\pi}{3}a_X^{-1} = \frac{4\pi\hbar^2}{(2M)}a_s^{(0)},$$

(46)

We therefore find that, $a_s^{(0)} = (13/3)a_X$. Thus, the ratio between the exact result and the Born approximation result for the exciton-exciton scattering length is about,

$$\left[\frac{a_s^{(0)}}{a_s}\right]_{\text{Coulomb}} = \frac{13/3}{1.5} \approx 2.89. \quad (47)$$

On the other hand, if we consider a contact interaction, the exact exciton-exciton scattering length in 3D is $0.6a_F$ [24] and the Born approximation result is $2a_F$, where $a_F$ is the fermion-fermion scattering length in 3D, and we find that,

$$\left[\frac{a_s^{(0)}}{a_s}\right]_{\text{contact}} = \frac{2}{0.6} \approx 3.33. \quad (48)$$

The two ratios are surprisingly close, despite the entirely different interaction potential between electrons and holes. This observation may suggest that the reduction in the exciton-exciton interaction strength or polariton-polariton interaction strength due to the beyond-Born-approximation effect could be universal, depending weakly on the underlying interaction between electrons and holes. We may then have a good reason to apply our toy model results with a contact interaction.

Therefore, it seems reasonable to consider a universal ratio defined by,

$$\frac{g_{PP}}{X_{LP}^{(0)}g_{XX}} = \left(\frac{\xi_{LP}}{X_{LP}}\right)F_{BB}, \quad (49)$$

which characterizes the two corrections: (i) the strong renormalization to $X_{LP}^4$ due to a very strong light-matter coupling within the Born approximation and (ii) the effect beyond the Born approximation. In Fig. 6 we report the inverse of this ratio as a function of the photon detuning at the light-matter coupling $\Omega = 0.8E_X$, at which the experimental data are taken. It is about 3 or 4 upon changing the photon detuning. The most contribution comes from the beyond-Born-approximation effect, as shown in the inset, which gives about a factor of 2 or 3 reduction to the polariton-polariton interaction strength.

We can now multiply the ratio $(\xi_{LP}^4 F_{BB}/X_{LP}^4)$ to the quasi-2D exciton-exciton interaction strength $g_{XX,\text{q2d}}^{(0)}$ in Eq. (7), to obtain a reasonable estimate for the polariton-polariton interaction strength. This is shown in Fig. 7 using a red solid line, together with the experimental data (blue dots with error bar) and the Born approximation result $g_{PP}^{(0)} = X_{LP}^{(0)}g_{XX}^{(0)}$ that was previously used as a theoretical upper bound (black dashed line). By taking into account the factor of 3 or 4 reduction, our beyond-Born-approximation theory seems to be in a reasonable agreement with the experimental data.

VI. CONCLUSIONS AND OUTLOOKS

In conclusions, we have theoretically investigated the beyond-Born-approximation effect for the polariton-
polariton interaction based on a Gaussian pair fluctuation theory [26], by using a toy model Hamiltonian with a contact interaction for electrons and holes. This simplified toy model enables us to understand the appearance of a constant polariton-polariton interaction strength, which is usually assumed in previous studies but is not theoretically guaranteed following the picture of a weakly interacting two-dimensional Bose gas of exciton-polaritons. We have shown that the effect beyond the Born approximation can lead to a factor of 3 reduction in the polariton-polariton interaction strength. As a by-product, the simplification also allows us to analytically define a generalized exciton Hopfield coefficient, Eq. (37), which takes into account the correction to the polariton-polariton interactions at large light-matter coupling. We have made an attempt to use our beyond-Born-approximation theory to understand the latest experimental data of the polariton-polariton interaction based on a Gaussian pair fluctuation approach [51, 52].

This agreement, however, should not be taken too seriously, as there are several improvements needed, both on the experimental side and theoretical side. In the experiments, the quantum well in microcavities is not in a perfect 2D configuration, in the sense that the width of the quantum well $l_z$ is comparable to the exciton Bohr radius $a_X$. The estimation of the quasi-2D Born approximation result in Eq. (7) is very rough. Furthermore, experimentally, in order to have very strong light-matter coupling, multiple isolated quantum wells were used and the electron-hole pairs in different quantum wells are all coupled to a single cavity photon field. This is somehow different from our theoretical model, which assumes a single quantum well in the microcavity.

Theoretically, on the other hand, we need to work out the exciton-exciton and polariton-polariton interaction strengths under the Coulomb-like interaction Eq. (11). The results within the Born approximation should be easy to obtain. We may simply generalize the work by Levinsen and his collaborators [22], paying specific attention to the renormalization of the light-matter coupling, as the exciton wave-functions are no longer analytically available. Going beyond the Born approximation will be very challenging. But, for the exciton-exciton interaction strength, at least we may try solving the four-particle problem (two electrons and two holes) in a numerically efficient way, using either fixed-node Monte Carlo simulation as in three dimensions [54] or explicitly correlated Gaussian basis expansion approach [51, 52].

ACKNOWLEDGMENTS

We thank Elena Ostrovskaya, Eliezer Estrecho, Maciej Pieczarka, Jesper Levinsen, Meera Parish and Jia Wang for helpful discussions. This research was supported by the Australian Research Council’s (ARC) Discovery Program, Grant No. DP170104008 (H.H.) and Grant No. DP180102018 (X.-J.L.), and by the Army Research Office under Awards W911NF-17-1-0312 (H.D.).

Appendix A: Generalized exciton Hopfield coefficient

We may clarify the physical meaning of the generalized exciton Hopfield coefficient from the electron-hole pair vertex function in vacuum $\Gamma_{\text{vac}}(Q = (q, i\nu_n))$, which takes the form,

$$\Gamma_{\text{vac}}(Q) = \left[\frac{1}{u_{\text{eff}}(Q)} + \Pi_{\text{vac}}(Q)\right]^{-1},$$

(A1)

$$\simeq C \frac{|\xi_{LP}(q)|^2}{i\nu_n - E_{LP}(q)}.$$  

(A2)

The second equation in the above holds near the pole $i\nu_n \rightarrow E_{LP}(q)$, with the constant $C$ and the generalized exciton Hopfield coefficient $\xi_{LP}^2(q)$ to be determined. Let us focus on the case $q = 0$ and recall that,

$$\frac{1}{u_{\text{eff}}(q = 0, i\nu_n)} + \Pi_{\text{vac}}(q = 0, i\nu_n) = \left(u + \frac{g^2}{i\nu_n - \delta}\right)^{-1} - \frac{M}{4\pi\hbar^2} \ln \left(\frac{-i\nu_n}{\varepsilon_0}\right).$$

(A3)

By Taylor-expanding the right-hand-side of the above equation in terms of the small quantity $x = i\nu_n - E_{LP}$, we find that,

$$\frac{1}{u_{\text{eff}}(q = 0, i\nu_n)} + \Pi_{\text{vac}}(q = 0, i\nu_n) \simeq \left[\frac{g^2/((\delta - E_{LP})^2}{(u + g^2/((\delta - E_{LP}))^2 - \frac{M}{4\pi\hbar^2} E_{LP} \right] (i\nu_n - E_{LP}).$$

(A4)

Therefore, we obtain

$$\Gamma_{\text{vac}}(q = 0, i\nu_n) \simeq \frac{4\pi\hbar^2}{M} (-E_{LP}) \frac{1}{i\nu_n - E_{LP}} \left[1 - \frac{4\pi\hbar^2}{Mu_{LP}^2 (\delta - E_{LP})^2}\right]^{-1},$$

(A5)
implying

\[ C = \frac{4\pi \hbar^2}{M} (-E_{LP}), \]

(A6)

\[ \xi_{LP}^2 (q = 0) = \left[ 1 - \frac{4\pi \hbar^2}{M u_L^2} \frac{g^2 E_{LP}}{\left( \delta - E_{LP} \right)^2} \right]^{-1}. \]

(A7)

**Appendix B: Density dependence of the ratio \( n_{\text{tot}}/n_{\text{MF}} \)**

Here we discuss the ratio \( n_{\text{tot}}/n_{\text{MF}} \) in the low-density limit. As shown in Fig. 8 upon decreasing total carrier density \( n_{\text{tot}} \) (or effectively bosonic chemical potential \( \mu_B \)), the ratio seems to saturate to a fixed value, which depends on the light-matter coupling \( \Omega \) and the photon detuning \( \delta \).

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FIG. 8. The ratio \( n_{\text{tot}}/n_{\text{MF}} \) as a function of the total carrier density, at a very strong light-matter coupling \( \Omega = 0.8E_X \) and at zero photon detuning \( \delta = 0 \).

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