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

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(Received 16 September 2020; accepted 19 November 2020; published 4 December 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 even in two dimensions, which result from coupling to the photon field. We show that quantum fluctuations lead to about a factor of 2 reduction in the polariton-polariton interaction strength within our toy model. Together with corrections to the 1*s* exciton approximation at very strong light-matter coupling, we find the polariton-polariton interaction strength under typical experimental conditions is overestimated by a factor of 3 in the widely used theories, if our toy model can qualitatively simulate the polariton interaction in GaAs quantum wells. 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 Estrecho *et al.* [Phys. Rev. B **100**, 035306 (2019)].

DOI: 10.1103/PhysRevA.102.063305

I. INTRODUCTION

Exciton-polaritons in microcavities are half-light and half-matter bosonic quasiparticles, arising from the strong coupling between the photo field and tightly-bound electron-hole pairs (i.e., excitons) [1,2]. Due to the ultra-small effective mass inherent from the light, Bose-Einstein condensation (BEC) of exciton-polaritons can occur at high temperatures [3,4]. Together with the nonlinearity originating from their underlying ferminoic constituents, exciton-polaritons provide an attractive platform to realize new technologies such as efficient and ultrafast optical switches and optical transistors [5,6].

Due to the critical role of the polariton nonlinearity in phase transitions and nonlinear optical device concepts, there have been intense experimental [7–14] and theoretical effort [15–19] to characterize the polariton nonlinearity over the past few decades. However, there continues to be conceptual difficulties in understanding and calculating the polariton nonlinearity with the widely used mean-field approach. The mean-field approach produces a constant polariton-polariton interaction strength $g_{\rm PP}$, or, a linearly increasing interaction energy with polariton density. This linear density dependence, however, is not anticipated for weakly interacting two-dimensional (2D) Bose gases [20]. According to the Bogoliubov theory, the relation between the chemical potential μ_B and the number density n of an interacting 2D Bose gas would be given by [21,22]

$$n \simeq \frac{m_B \mu_B}{4\pi\hbar^2} \ln\left(\frac{4\hbar^2}{m_B \mu_B a_s^2 e^{2\gamma+1}}\right), \tag{1}$$

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 [20], and $\gamma \simeq 0.577$ is Euler's constant. This indicates a density or chemical potential dependent interaction strength

$$g(\mu_B) = \frac{\mu_B}{n} = \frac{4\pi\hbar^2}{m_B} \ln^{-1}\left(\frac{4\hbar^2}{m_B \mu_B a_s^2 e^{2\gamma+1}}\right). \tag{2}$$

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, if we treat polaritons as a gas of weakly interacting bosons. In greater detail, to date most calculations of the polariton-polariton interaction strength are based on the Born approximation [23]. In the exciton-polariton model, it leads to a polariton-polariton interaction strength [16]

$$g_{\rm PP}^{(0)} = X_{\rm LP}^4 g_{XX}^{(0)}, \tag{3}$$

where

$$g_{XX}^{(0)} \simeq 6.06 E_X a_X^2 = 6.06\hbar^2/M \tag{4}$$

is the constant exciton-exciton interaction strength in 2D and

$$X_{\rm LP}^2 = \frac{1}{2}\left(1 + \frac{\delta/2}{\sqrt{\delta^2/4 + \Omega^2}}\right) \tag{5}$$

is the excitonic Hopfield coefficient with the photon detuning δ (measured with respect to the exciton energy $-E_X$) and with the light-matter coupling Ω. Here, a_X and $E_X = \hbar^2/(M a_X^2)$ are the Bohr radius and binding energy of excitons, respectively, and we assumed for simplicity that electrons and holes take the same mass $m_\mathrm{e} = m_\mathrm{h} = m_\mathrm{eh} = M$. We also used the superscript "0" to explicitly indicate the results within the Born approximation. Equation (3) 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 Ω becomes strong and comparable to E_X, so that the standard exciton-polariton model starts to break down. This nontrivial effect due to strong light-matter coupling is well known in the literature [8,16] 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 [19]. It was shown that the correction to Eq. (3) can be about $\sim$20% at the very strong coupling regime when $\Omega \sim E_X$ [19]. On the other hand, experimentally, the exciton-exciton interaction strength $g_{XX}^{(0)}$ in Eq. (3) may also need revision, considering the quasi-2D configuration of the quantum well, whose width l_z would be similar to a_X [13]. In such a situation, a rough estimation gives rise to

$$g_{XX,\mathrm{q2d}}^{(0)} = \frac{26\pi}{3} E_X a_X^2 \left(\frac{a_X}{l_z} \right). \tag{6}$$

This expression was used by Estrecho and his collaborators to set a theoretical *upper* bound for the polariton-polariton interaction strength [13]. 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 two dimensions, where quantum and thermal fluctuations are so significant that the equation of state of the system can qualitatively be altered [24]. 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 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 three-dimensional (3D) s-wave length length a_F. In the BEC limit where tightly bound molecules are formed, the exact molecule-molecule scattering length is $a_s \simeq 0.6 a_F$ [25,26], 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 two dimensions 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 of ultracold atoms [27,28], we reliably calculate the polariton-polariton interaction strength at various light-matter couplings and photon detunings for the toy model.

Two main observations are worth noting. First, in the presence of the photon field, the scattering of two composite bosons (i.e., excitons) is strongly modified. In particular, at strong light-matter coupling, where the photon field is notably populated, the internal fermionic degree of freedom of excitons cannot be ignored. The modification to the exciton-exciton scattering due to the photon field provides the correct theoretical understanding why a nearly constant, density-independent polariton-polariton interaction strength was found in the experiments [13]. Second, the effect beyond the Born approximation is significant and typically leads to about a factor of 2 reduction in the interaction strength. Combined with the nontrivial effect due to strong light-matter coupling, in total we find that the polariton-polariton interaction strength under typical experimental conditions to be about a factor of 3 smaller relative to the prediction of Eq. (3).

We note that, for a small light-matter coupling, we may use a purely bosonic model Hamiltonian to describe the exciton-polariton system [29]. In that case, the beyond mean-field effect can be captured by using the Bogoliubov theory, which takes into account the many-body effects and strong quantum fluctuations in two dimensions [29], and momentum-dependent interactions may also be used without the simplification to contact interactions. Our GPF results from the fermionic toy model agree well with the analytic Bogoliubov predictions obtained with the bosonic exciton-polariton model, if we use the same parameters under the same condition.

The rest of the paper is organized as follows. In Sec. II, we briefly review the GPF theory of the toy model with a contact electron-hole interaction 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 [19]. 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 attraction and compare our prediction with the latest measurement of the polariton-polariton interaction strength [13]. 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 $\mathscr{H} = \mathscr{H}_0 + \mathscr{H}_{\mathrm{LM}} + \mathscr{H}_\mathrm{C}$ as [30–32]

$$\mathscr{H}_0 = \sum_{\mathbf{k}\sigma} \xi_\mathbf{k} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \sum_\mathbf{q} \left(\frac{\hbar^2 \mathbf{q}^2}{2m_\mathrm{ph}} + \delta_0 - \mu \right) \phi_\mathbf{q}^\dagger \phi_\mathbf{q}, \tag{7}$$

$$\mathscr{H}_{\mathrm{LM}} = \frac{g_0}{\sqrt{\mathcal{S}}} \sum_{\mathbf{kq}} \left(\phi_\mathbf{q}^\dagger c_{\frac{\mathbf{q}}{2}-\mathbf{k}h} c_{\frac{\mathbf{q}}{2}+\mathbf{k}e} + \mathrm{H.c.} \right), \tag{8}$$

$$\mathscr{H}_\mathrm{C} = \frac{1}{2\mathcal{S}} \sum_{\mathbf{kk}'\mathbf{q}}^{\sigma\sigma'} V_{\mathbf{kk}'}^{\sigma\sigma'} c_{\frac{\mathbf{q}}{2}+\mathbf{k}\sigma}^\dagger c_{\frac{\mathbf{q}}{2}-\mathbf{k}\sigma'}^\dagger c_{\frac{\mathbf{q}}{2}-\mathbf{k}'\sigma'} c_{\frac{\mathbf{q}}{2}+\mathbf{k}'\sigma}. \tag{9}$$

Here, $\xi_{\mathbf{k}} \equiv \hbar^2\mathbf{k}^2/(2M) - \mu/2$, δ_0, μ, g_0 and $\mathcal{S}$ 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 $M = m_{\rm eh} \simeq 0.067m_0$ for electrons and holes (where m_0 is the free-electron mass) and an ultra-small photonic mass $m_{\rm ph} \simeq 3 \times 10^{-5}m_0$ due to the microcavity confinement [1]. $c_{\mathbf{k}\sigma}$ are the annihilation operators of electrons ($\sigma = e$) and holes ($\sigma = h$), and $\phi_{\mathbf{q}}$ denote the annihilation operators of photons.

In Eq. (9), $V_{\mathbf{kk}'}^{\sigma\sigma'}$ are the Coulomb-like interactions among electrons and holes, and are defined as the Fourier transformation of a screened potential [33,34]

$$V_C^{\sigma\sigma'}(r) = \chi_{\sigma\sigma'}\frac{\pi e^2}{2\varepsilon_s r_0}\left[H_0\left(\frac{r}{r_0}\right) - Y_0\left(\frac{r}{r_0}\right)\right], \qquad (10)$$

where $\chi_{\sigma\sigma'} = +1$ for $\sigma = \sigma'$ and $\chi_{\sigma\sigma'} = -1$ for $\sigma \neq \sigma'$, ε_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 [33,34]. The model Hamiltonian is extremely difficult to solve because of the *nonlocal* nature of the Coulomb interaction. To find a way around this, we propose a *toy* model by replacing the Coulomb interaction with a local contact interaction [32,35], i.e.,

$$\mathscr{H}_{\rm C} = \frac{u_0}{\mathcal{S}}\sum_{\mathbf{kk}'\mathbf{q}} c^\dagger_{\frac{\mathbf{q}}{2}+\mathbf{k}e} c^\dagger_{\frac{\mathbf{q}}{2}-\mathbf{k}h} c_{\frac{\mathbf{q}}{2}-\mathbf{k}'h} c_{\frac{\mathbf{q}}{2}+\mathbf{k}'e}, \qquad (11)$$

where the interaction strength u_0 should be tuned to reproduce the correct ground-state energy of excitons with the Coulomb-like interaction Eq. (10).

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 [36–39]. 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 $m_{\rm ph} = 2M$, which holds for ultracold atoms), while the excitons at low density correspond to the tightly bound Cooper pairs in the open channel. For more details, we refer to the discussions in Refs. [27,32]. At a broad Feshbach resonance, which is realized when the light-matter coupling is *infinitely* strong, our toy model Hamiltonian has actually been investigated both experimentally [40–42] and theoretically [43,44]. Here, the purpose of this work is to understand the molecular scattering 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 δ_0 will be replaced by u, g, and $\tilde{\delta} = \delta - E_X = E_{\rm cav}$, respectively. Here, $E_{\rm cav}$ is the cavity energy measured from the edge of the band gap, 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 Ω as follows [27]:

$$u = \frac{4\pi\hbar^2}{M}\ln^{-1}\left(\frac{E_X}{\varepsilon_0}\right), \qquad (12)$$

$$g = 2\sqrt{\pi}\Omega a_X \ln^{-1}\left(\frac{E_X}{\varepsilon_0}\right), \qquad (13)$$

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 the Supplemental Material of Ref. [27], 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 [24,45–47] and then consider the low-density dilute limit. The details of the GPF formalism are again outlined in Ref. [27]. Here, for self-containedness we briefly review the main equations. Taking the Hubbard-Stratonovich transformation, we first introduce a pairing field to decouple $\mathscr{H}_{\rm C}$ in Eq. (11) and integrate out the fermionic fields $c_{\mathbf{k}\sigma}$. 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 [27]

$$\Omega_{\rm MF} = -\frac{\Delta^2}{u_{\rm eff}} + \sum_{\mathbf{k}}\left(\xi_{\mathbf{k}} - E_{\mathbf{k}} + \frac{\Delta^2}{\hbar^2\mathbf{k}^2/M + \varepsilon_0}\right), \qquad (14)$$

where Δ is an order parameter satisfying the gap equation $\partial\Omega_{\rm MF}/\partial\Delta = 0$,

$$u_{\rm eff} \equiv u + \frac{g^2}{\mu - \tilde{\delta}} \qquad (15)$$

is an effective interaction incorporating the photon-mediated attraction, and $E_{\mathbf{k}} \equiv \sqrt{\xi_{\mathbf{k}}^2 + \Delta^2}$ is the dispersion relation for fermionic Bogoliubov quasiparticles. To go beyond mean field, we expand the effective action around the saddle point and keep the bilinear terms in the polariton field (i.e., the so-called Gaussian fluctuations) [45–48]. Integrating out these fluctuations, we obtain the GPF thermodynamic potential from quantum fluctuations [27]

$$\Omega_{\rm GPF} = \frac{1}{2}k_BT\sum_{\mathcal{Q}}\ln\det\Gamma[\mathcal{Q} = (\mathbf{q}, i\nu_n)]e^{i\nu_n 0^+}, \qquad (16)$$

where $\Gamma(\mathcal{Q})$ with bosonic Matsubara frequencies $\nu_n = 2\pi nk_BT$ ($n \in \mathbb{Z}$) is the Green's function of the polariton field. It is a 2×2 matrix with off-diagonal terms representing the phase correlation of the superfluid. In the normal phase above the superfluid transition temperature, the off-diagonal terms disappear and the diagonal term becomes a scalar variable [27]

$$\Gamma(\mathcal{Q}) = \left[\frac{1}{u_{\rm eff}(\mathcal{Q})} + \Pi(\mathcal{Q})\right]^{-1}, \qquad (17)$$

where $u_{\mathrm{eff}}(Q) \equiv u + g^2/[i\nu_n - \hbar^2\mathbf{q}^2/(2m_{\mathrm{ph}}) + \mu - \tilde{\delta}]$ is a momentum- and frequency-dependent effective interaction strength, and $\Pi(Q)$ is the pair propagator. In the vacuum limit (i.e., the two-particle limit), the pair propagator takes the form [27]

$$\Pi_{\mathrm{vac}}(Q) = -\frac{M}{4\pi\hbar^2}\ln\left(\frac{\hbar^2 q^2/(4M) - i\nu_n}{\varepsilon_0}\right). \quad (18)$$

By substituting the above vacuum pair propagator into Eq. (17), we can determine the pole of the polariton Green's function and obtain the dispersion relation of the polaritons in the dilute limit, which consists of two branches: the lower-polariton branch $E_{\mathrm{LP}}(\mathbf{q})$ and the upper-polariton branch $E_{\mathrm{UP}}(\mathbf{q})$ [27].

The GPF theory of exciton-polaritons is easy to numerically implement. For a given chemical potential, we determine the order parameter using the gap equation. The mean-field and GPF thermodynamic potentials are then calculated, from which we obtain the total carrier densities $n_{\mathrm{tot}} = n_{\mathrm{MF}} + n_{\mathrm{GPF}}$, where

$$n_{\mathrm{MF}} = -\frac{\partial\Omega_{\mathrm{MF}}}{\partial\mu}, \quad (19)$$

$$n_{\mathrm{GPF}} = -\frac{\partial\Omega_{\mathrm{GPF}}}{\partial\mu}. \quad (20)$$

One advantage of our GPF theory is that it can provide a reliable equation of state at zero temperature [24,45,47]. In particular, in the dilute limit, where the chemical potential depends linearly on the density (i.e., the linear regime), it gives an approximate but reasonably accurate molecular scattering length. For example, for a two-component interacting Fermi gas at the BEC-BCS crossover in three dimensions, the molecular scattering length predicted by the GPF theory is about $a_s \simeq 0.55a_F$ [45,47], which is slightly smaller than the exact value $a_s \simeq 0.60a_F$ [25]. In two dimensions of interest, the GPF theory also provides a very accurate molecular scattering length [24], as we shall discuss in detail in the next section.

III. 2D EXCITON CONDENSATE WITH CONTACT INTERACTIONS

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 [24,40], 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 is related to the 2D scattering length between fermions $a_{2\mathrm{D}}$ through [49]

$$a_s = \kappa a_{2\mathrm{D}} \simeq 0.56 a_{2\mathrm{D}}. \quad (21)$$

Here, $a_{2\mathrm{D}} = 2e^{-\gamma}a_X$ can be calculated by using the binding energy $E_X = 4\hbar^2/(Ma_{2\mathrm{D}}^2 e^{2\gamma})$. The molecular scattering length determined from the 2D GPF theory *coincides*with the exact value if we keep the two significant digits [24]. According to the Bogoliubov theory of a 2D weakly interacting Bose gas, Eq. (1), we thus obtain

$$n \simeq \frac{1}{2\pi a_X^2}\left[-2\ln\kappa - (\ln 2 + 1) - \ln\frac{\mu_B}{E_X}\right]\frac{\mu_B}{E_X}. \quad (22)$$

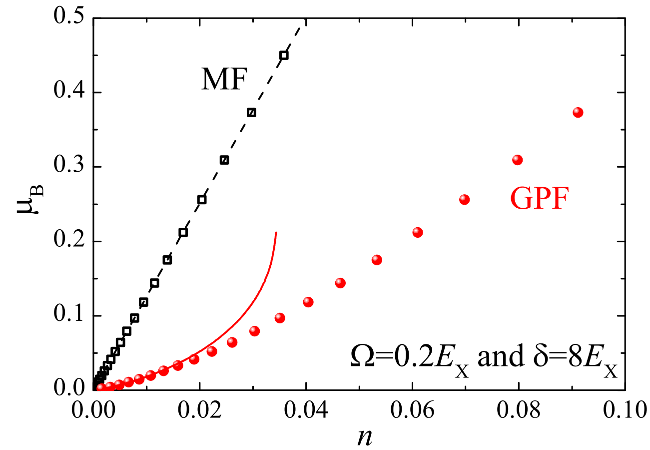

FIG. 1. Bosonic chemical potential (in units of E_X) as a function of the number density (in units of a_X^{-2}) at strong light-matter coupling $\Omega = 0.2E_X$ and at a large photon detuning $\delta = 8E_X$. The black empty squares and red solid circles show the results obtained by mean-field and Gaussian pair fluctuation theories, respectively. The black dashed line and blue solid line are the predictions of the mean-field theory, Eq. (24), and Bogoliubov theory for excitons, Eq. (22), in the dilute density limit, i.e., $y = (4\pi)x$ and $y = 2\pi x/[-2\ln\kappa - (\ln 2 + 1) - \ln x] \simeq 2\pi x/(-0.5335 - \ln x)$, where $x \equiv na_X^2$ and $y \equiv \mu_B/E_X$. At the density $n > 0.02a_X^{-2}$, the GPF results cannot be explained by the Bogoliubov theory. This is anticipated since the gas parameter $na_X^2 > 0.02$ is already too large and the system is no longer in the weakly interacting regime.

In contrast, the mean-field theory cannot predict qualitatively correct equation of state. By writing the molecular chemical potential μ_B in terms of the chemical potential of fermions $\mu_F = \mu/2$ (i.e., $\mu_B = 2\mu_F + E_X = \mu + E_X$), from the mean-field equation of state [24]

$$\mu_F + \frac{E_X}{2} = \varepsilon_F \equiv \frac{\hbar^2(2\pi n_F)}{2M}, \quad (23)$$

we find that

$$\mu_B = \frac{4\pi\hbar^2 n}{M} = 4\pi E_X a_X^2 n, \quad (24)$$

implying a molecule-molecule interaction strength $g_m = 4\pi E_X a_X^2$ 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

$$g_{XX}^{(0)} = 4\pi E_X a_X^2, \quad (25)$$

which is about two times the exciton-exciton interaction strength in Eq. (4) 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 $n = n_{\mathrm{tot}}$ or small chemical potential $\mu_B = \mu - E_{\mathrm{LP}}$, where $E_{\mathrm{LP}} = -E_X$ is the energy of the zero-momentum

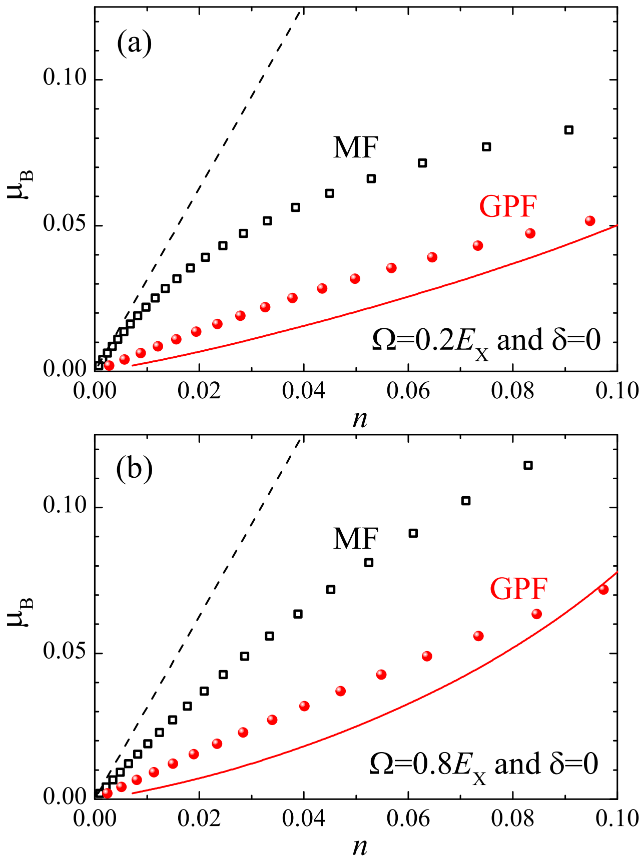

FIG. 2. Bosonic chemical potential (in units of E_X) as a function of the number density (in units of a_X^{-2}) with zero photon detuning $\delta = 0$, at (a) strong light-matter coupling $\Omega = 0.2E_X$ and (b) at very strong light-matter coupling $\Omega = 0.8E_X$. The black empty squares and red solid circles show the results obtained by mean-field and Gaussian pair fluctuation theories, respectively. The black dashed line is the result from the Born approximation $g_{PP}^{(0)} = X_{LP}^4(4\pi E_X a_X^2)$. The red solid line is based on the anticipation of a weakly interacting 2D Bose gas of exciton-polaritons, i.e., $g_{PP} = X_{LP}^4 g_{XX}$, where g_{XX} is give by Eq. (26). Here, $X_{LP}^2 = 1/2$ at zero detuning according to the exciton-polariton model Eq. (5).

lower-polariton 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 Eqs. (24) and (22), respectively. We emphasize that, within the GPF theory, the chemical potential dependent exciton-exciton interaction strength is given by

$$g_{XX}(\mu_B) = \frac{2\pi E_X a_X^2}{-2\ln\kappa - (\ln 2 + 1) - \ln(\mu_B/E_X)}. \tag{26}$$

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 at two light-matter couplings $\Omega = 0.2E_X$ [Fig. 2(a)] and $\Omega = 0.8E_X$ [Fig. 2(b)]. For comparison, we show also the corresponding equations of state predicted by the exciton-polariton model [see, i.e., Eq. (3)] 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 $g_{\rm PP}^{(0)} = X_{\rm LP}^4 g_{XX}^{(0)}$ [16], 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.

A. Born approximation (mean field)

Let us first analyze the mean-field results. From the mean-field thermodynamic potential Eq. (14), we may derive the gap equation

$$\sqrt{\mu^2 + 4\Delta^2} - \mu = 2\varepsilon_0 \exp\left(\frac{4\pi\hbar^2}{M u_{\rm eff}}\right), \tag{27}$$

and the number equation

$$n_{\rm MF} = \left(\frac{g}{\tilde{\delta} - \mu}\right)^2 \frac{\Delta^2}{u_{\rm eff}^2} + \frac{M}{8\pi\hbar^2}\left(\sqrt{\mu^2 + 4\Delta^2} + \mu\right). \tag{28}$$

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

$$u_{\rm eff} \to u + \frac{g^2}{E_{\rm LP} - \tilde{\delta}} \equiv u_{\rm LP}. \tag{29}$$

Taylor-expanding the gap equation, we find that

$$-E_{\rm LP} - \mu_B - \frac{\Delta^2}{E_{\rm LP}} = \varepsilon_0 \exp\left(\frac{4\pi\hbar^2}{M u_{\rm LP}}\right)\left[1 + \frac{\mu_B}{\mathcal{A}}\right], \tag{30}$$

where

$$\mathcal{A}^{-1} \equiv \frac{4\pi\hbar^2}{M u_{\rm LP}^2} \frac{g^2}{(\tilde{\delta} - E_{\rm LP})^2}. \tag{31}$$

The leading term of the above gap equation is simply the expression for the zero-momentum lower-polariton energy [27], i.e., $E_{\rm LP} = -\varepsilon_0 e^{4\pi\hbar^2/(M u_{\rm LP})}$. Using this to eliminate the cutoff energy scale ε_0, we obtain

$$-\frac{\Delta^2}{E_{\rm LP}} = \left[1 - \frac{E_{\rm LP}}{\mathcal{A}}\right]\mu_B. \tag{32}$$

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

$$n_{\rm MF} = \frac{M}{4\pi\hbar^2}\left[1 - \frac{E_{\rm LP}}{\mathcal{A}}\right]\left(-\frac{\Delta^2}{E_{\rm LP}}\right). \tag{33}$$

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

$$\mu_B = \left[1 - \frac{E_{\rm LP}}{\mathcal{A}}\right]^{-2} (4\pi E_X a_X^2) n. \tag{34}$$

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

$$g_{PP}^{(0)} = \xi_{\rm LP}^4 \left(4\pi E_X a_X^2\right), \tag{35}$$

where we defined

$$\xi_{\rm LP}^2 \equiv \left[1 - \frac{E_{\rm LP}}{\mathcal{A}}\right]^{-1} = \left[1 - \frac{4\pi\hbar^2}{Mu_{\rm LP}^2}\frac{g^2 E_{\rm LP}}{(\tilde{\delta} - E_{\rm LP})^2}\right]^{-1}. \tag{36}$$

By recalling that $4\pi E_X a_X^2 = g_{XX}^{(0)}$ is the exciton-exciton interaction strength for our toy model, we may interpret $\xi_{\rm LP}^2$ 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 $u_{\rm LP} \simeq u$ and use the expression for the zero-momentum lower-polariton energy

$$E_{\rm LP} = -E_X + \frac{\delta}{2} - \sqrt{\frac{\delta^2}{4} + \Omega^2}. \tag{37}$$

By further taking $g^2/u^2 = M\Omega^2/(4\pi\hbar^2 E_X)$ and recalling that $\delta = \tilde{\delta} + E_X$, we find that

$$\xi_{\rm LP}^2 \simeq \left[1 + \frac{\Omega^2}{(\tilde{\delta} - E_{\rm LP})^2}\right]^{-1} = X_{\rm LP}^2. \tag{38}$$

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

In Fig. 3, we report the Hopfield coefficients $X_{\rm LP}^2$ (black dashed line) and $\xi_{\rm LP}^2$ (red solid line) as a function of the photon detuning at three light-matter couplings: $\Omega = 0.1E_X$ [Fig. 3(a)], $\Omega = 0.5E_X$ [Fig. 3(b)], and $\Omega = 1.0E_X$ [Fig. 3(c)]. At small coupling $\Omega \ll E_X$ as shown in Fig. 3(a), $\xi_{\rm LP}^2$ is essentially the same as the $X_{\rm LP}^2$, as we already confirmed analytically. However, as the light-matter coupling increases, $\xi_{\rm LP}^2$ becomes increasingly smaller than $X_{\rm LP}^2$ 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 $\xi_{\rm LP}^2$ and $X_{\rm LP}^2$ 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 Ω/E_X was derived analytically [8,16]. The saturation correction *enhances* the polariton-polariton interaction strength. This difference was also numerically investigated by Levinsen and coworkers most recently [19]. 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 [50]. 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 [19]. 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 $\xi_{\rm LP}^2$ in comparison with $X_{\rm LP}^2$.

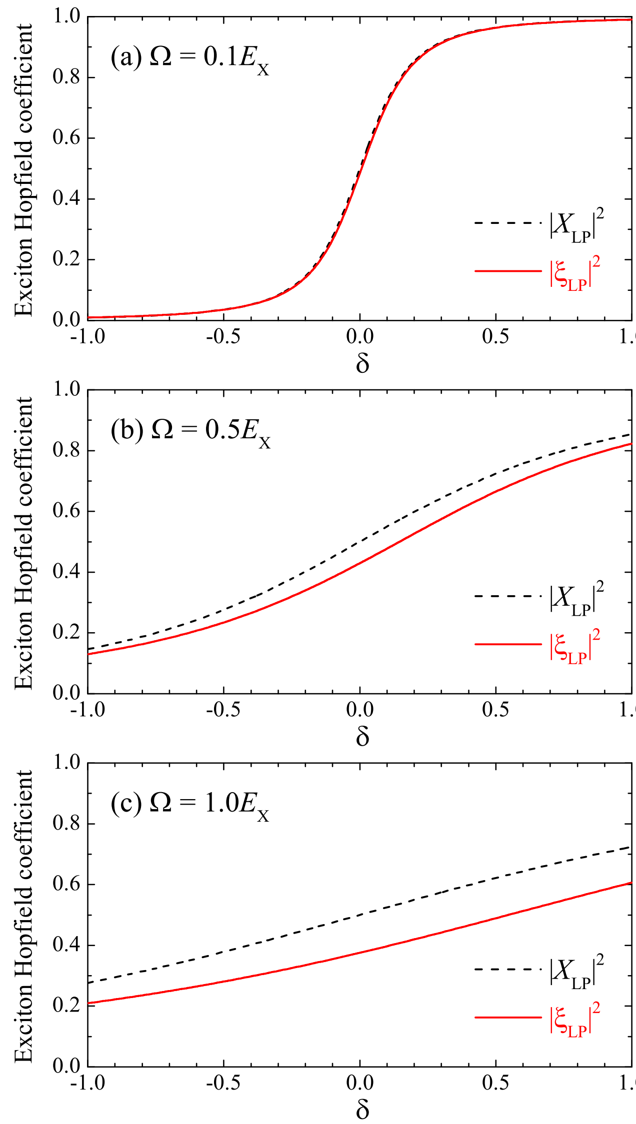

FIG. 3. Exciton Hopfield coefficient $X_{\rm LP}^2$ (black dashed line) and the generalized exciton Hopfield coefficient $\xi_{\rm LP}^2$ (red solid line) as a function of the photon detuning δ at three light-matter couplings: $\Omega =$ (a) $0.1E_X$, (b) $0.5E_X$, and (c) $1.0E_X$.

B. Beyond the Born approximation (GPF)

Here, we turn to consider the beyond-Born-approximation effect using the GPF theory. Naïvely, we argue that the polariton system consists of different types of carriers [27], as characterized by $n_{\rm MF}$ and $n_{\rm GPF}$, 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., $n_{\rm MF} \ll n_{\rm GPF}$, the system could be viewed as a weakly interacting Bose gas of exciton-polaritons and the density equation of state then follows the Bogoliubov theory, as we 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 $n_{\rm MF}$ and $n_{\rm GPF}$ become significant and towards the zero-density limit, their ratio $n_{\rm MF}/n_{\rm GPF}$ saturates to a constant. At large light-matter coupling and near

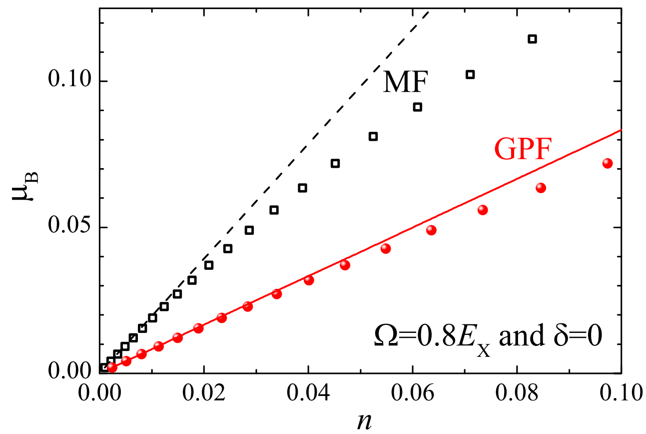

FIG. 4. Bosonic chemical potential (in units of E_X) as a function of the number density (in units of a_X^{-2}) with zero photon detuning $\delta = 0$ at very strong light-matter coupling $\Omega = 0.8E_X$. As the same as shown in Fig. 2(b), the black empty squares and red solid circles show the results obtained by mean-field and Gaussian pair fluctuation theories, respectively. But, now the black dashed line is the result from the Born approximation, $g_{\rm PP}^{(0)} = \xi_{\rm LP}^4(4\pi E_X a_X^2)$, with the generalized exciton Hopfield coefficient $\xi_{\rm LP}^2$. The red solid line shows the result $g_{\rm PP} = \xi_{\rm LP}^4 \mathcal{F}_{BB}(4\pi E_X a_X^2)$, which takes into account the reduction beyond the Born approximation.

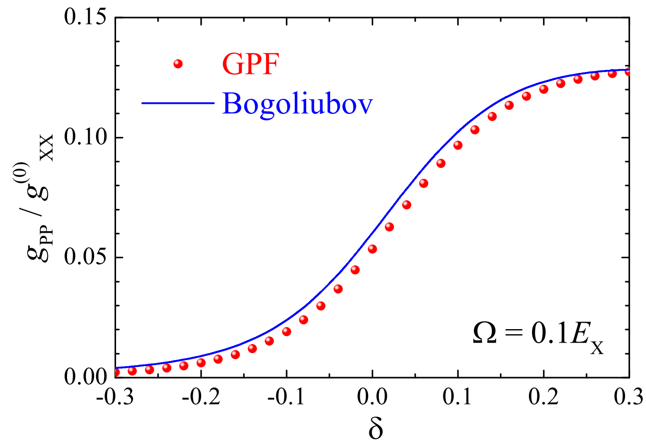

FIG. 5. The ratio $g_{\rm PP}/g_{XX}^{(0)}$ as a function of the photon detuning δ (in units of E_X) at a light-matter coupling $\Omega = 0.1E_X$. The GPF result (red circles) is compared with an analytic prediction from the exciton-polariton model within the Bogoliubov theory (blue line), Eq. (43).

zero photon detuning, therefore, we may define a quantity

$$\mathcal{F}_{BB} = \lim_{\mu_B \to 0} \left(\frac{n_{\rm MF}}{n_{\rm tot}} \right), \tag{39}$$

which itself is functions of the light-matter coupling Ω and of the photon detuning δ. Now, using Eqs. (32) and (33) for $n_{\rm MF}$, in the zero-density limit we find

$$\mu_B = 4\pi E_X a_X^2 \xi_{\rm LP}^4 n_{\rm MF} = \xi_{\rm LP}^4 \mathcal{F}_{BB}\left(4\pi E_X a_X^2\right) n, \tag{40}$$

which implies a polariton-polariton interaction strength

$$g_{PP} = \left(\xi_{\rm LP}^4 \mathcal{F}_{BB}\right)\left(4\pi E_X a_X^2\right). \tag{41}$$

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 $g_{\rm PP}^{(0)} = \xi_{\rm LP}^4(4\pi E_X a_X^2)$. 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 $g_{\rm PP}$ 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 this work,

$$\frac{g_{\rm PP}}{g_{XX}^{(0)}} = \xi_{\rm LP}^4 \mathcal{F}_{BB}, \tag{42}$$

where $\xi_{\rm LP}^4$ is responsible for the large light-matter coupling and $\mathcal{F}_{BB}$ accounts for the beyond-Born-approximation effect. In Fig. 4, we replot Fig. 2(b) and add the anticipated behavior Eq. (35) for the mean-field result (black dashed line) and Eq. (41) 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 [29] or the scattering theory [51]. 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 $\kappa \simeq 0.56$ as discussed in Sec. III) for a contact electron-hole attraction, it takes the form [29,51]

$$\frac{g_{\rm PP}}{g_{XX}^{(0)}} = \frac{X_{\rm LP}^4}{2\ln\left[E_X/\left|E_{\rm LP}^{(0)}\right|\right] - 4\ln(2\kappa)}, \tag{43}$$

where $E_{\rm LP}^{(0)} \equiv E_{\rm LP} - (-E_X) = \delta/2 - \sqrt{\delta^2/4 + \Omega^2} < 0$ is the energy of zero-momentum lower-polariton, measured with respect to the exciton energy $-E_X$. At small light-matter coupling, we have $\xi_{\rm LP}^2 = X_{\rm LP}^2$. Therefore, by comparing Eqs. (42) and (43), we obtain that for $\Omega \ll E_X$,

$$\mathcal{F}_{BB} = \frac{1}{2\ln\left[E_X/\left|E_{\rm LP}^{(0)}\right|\right] - 4\ln(2\kappa)}. \tag{44}$$

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 versus bosonic) are used, both of them reliably describe the exciton-polariton physics at small light-matter coupling.

It is interesting to note that Eq. (43) clearly shows a pole at the lower-polariton energy $E_{\rm LP}^{(0)} = -E_X/(4\kappa^2) \simeq$

$-0.8E_X$ or $\delta \simeq -0.8E_X$ under the condition $\Omega \ll E_X$. This *weak* logarithmic divergence is neutralized by the rapidly decreasing excitonic Hopfield coefficient $X_{\rm LP}^4 \simeq (\Omega/\delta)^4 \sim 2.4 \times 10^{-4}$, if we take $\Omega = 0.1E_X$. As a result, the polariton-polariton interaction strength $g_{\rm PP}$ is always much smaller than the exciton-exciton interaction strength $g_{XX}^{(0)}$ obtained within the Born approximation. This situation, however, can dramatically change if the ratio κ is allowed to tune experimentally (hopefully in transition-metal-dichalcogenide monolayers [29]). An enlarged ratio κ shifts the logarithmic pole in Eq. (43) to the zero photon detuning $\delta \sim 0$ and consequently the polariton-polariton interaction strength $g_{\rm PP}$ could be greatly enhanced. For more detailed discussions, we refer to Ref. [29].

V. COMPARISON TO THE EXPERIMENT

Although our main result Eq. (42) 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. (10), should be considered. To this aim, let us make a *bold* assumption that, Eq. (42) 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 three dimensions, 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 three dimensions [52], 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

$$g_{XX}^{(0)} = \frac{26\pi}{3} E_X a_X^3 \equiv \frac{4\pi\hbar^2}{(2M)} a_s^{(0)}, \tag{45}$$

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

$$\left[\frac{a_s^{(0)}}{a_s}\right]_{\rm Coulomb} = \frac{13/3}{1.5} \simeq 2.89. \tag{46}$$

On the other hand, if we consider a contact interaction, the exact exciton-exciton scattering length in three dimensions is $0.6a_F$ [25] and the Born approximation result is $2a_F$, where a_F is the fermion-fermion scattering length in three dimensions, and we find that

$$\left[\frac{a_s^{(0)}}{a_s}\right]_{\rm contact} = \frac{2}{0.6} \simeq 3.33. \tag{47}$$

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.

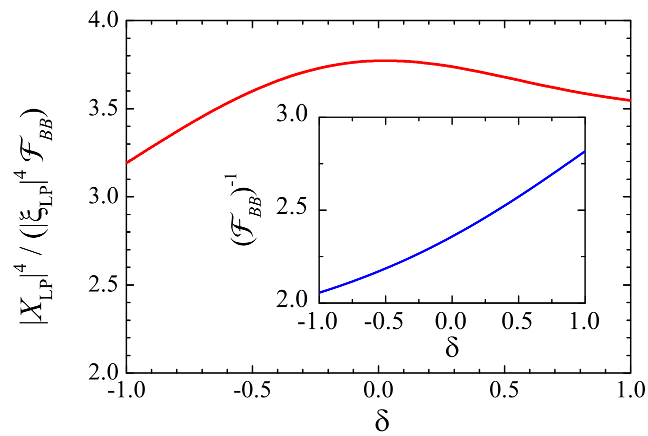

FIG. 6. The reduction factor in the polariton-polariton interaction strength at a very strong light-matter coupling $\Omega = 0.8E_X$ as a function of the photon detuning δ due to the combined effects of the saturation in exciton oscillator strength and the beyond-Born-approximation correction. The inset shows the inverse density fraction of fermionic quasi-particles, $\mathcal{F}_{BB}^{-1} = n_{\rm tot}/n_{\rm MF}$, as a function of the photon detuning δ.

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

$$\frac{g_{\rm PP}}{X_{\rm LP}^4 g_{XX}^{(0)}} = \left(\frac{\xi_{\rm LP}^4}{X_{\rm LP}^4}\right)\mathcal{F}_{BB}, \tag{48}$$

which characterizes the two corrections: (i) the strong renormalization to $X_{\rm LP}^4$ 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 $(\xi_{\rm LP}^4\mathcal{F}_{BB}/X_{\rm LP}^4)$ to the quasi-2D exciton-exciton interaction strength $g_{XX,\rm q2d}^{(0)}$ in Eq. (6), 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 $g_{PP}^{(0)} = X_{\rm LP}^4 g_{XX,\rm q2d}^{(0)}$ 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. CONCLUSION AND OUTLOOK

In conclusion, we theoretically investigated the beyond-Born-approximation effect for the polariton-polariton interaction based on a Gaussian pair fluctuation theory [27], by using a toy model Hamiltonian with a contact interaction

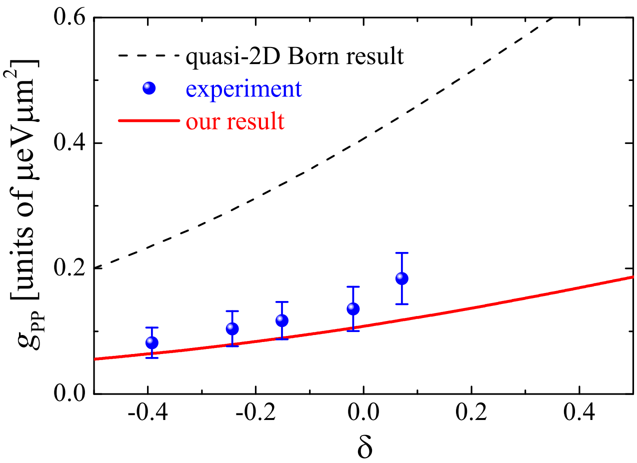

FIG. 7. Theory versus experiment for the polariton-polariton interaction strength at a very strong light-matter coupling $\Omega \simeq 0.8E_X$. Our beyond-Born-approximation prediction (red solid line) is compared with the experimental data (blue circles with error bars) that is taken from Fig. 5(a) in Ref. [13]. The black dashed line shows the result obtained with the Born approximation in a quasi-2D configuration, i.e., Eq. (6), together with X_{LP}^2 calculated using Eq. (5). Our beyond-Born-approximation prediction is calculated by dividing the quasi-2D Born approximation result by the reduction factor shown in Fig. 6. It takes into account both the saturation effect in the exciton oscillator strength and the correction beyond the Born approximation.

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 2D Bose gas of exciton-polaritons. We showed 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. (36), which takes into account the correction to the polariton-polariton interactions at large light-matter coupling. We made an attempt to use our beyond-Born-approximation theory to understand the latest experimental data of the polariton-polariton interaction strength [13]. A reasonable agreement has been found.

Future work will solve the exciton-exciton and polariton-polariton interaction strengths under the Coulomb-like interaction Eq. (10). The results within the Born approximation should be easy to obtain. We may simply generalize the work by Levinsen and his collaborators [19], 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 [52] or explicitly correlated Gaussian basis expansion approach [53,54].

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 National Science Foundation under Grant No. DMR 2004287 (H.D.) and by the Army Research Office under Award 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 $\Gamma_{vac}[\mathcal{Q} = (\mathbf{q}, i\nu_n)]$, which takes the form

$$\Gamma_{vac}(\mathcal{Q}) = \left[\frac{1}{u_{eff}(\mathcal{Q})} + \Pi_{vac}(\mathcal{Q})\right]^{-1}, \tag{A1}$$

$$\simeq C\frac{|\xi_{LP}(\mathbf{q})|^2}{i\nu_n - E_{LP}(\mathbf{q})}. \tag{A2}$$

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

$$\frac{1}{u_{eff}(\mathbf{q}=\mathbf{0}, i\nu_n)} + \Pi_{vac}(\mathbf{q}=\mathbf{0}, i\nu_n) = \left(u + \frac{g^2}{i\nu_n - \tilde{\delta}}\right)^{-1} - \frac{M}{4\pi\hbar^2}\ln\left(\frac{-i\nu_n}{\varepsilon_0}\right). \tag{A3}$$

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

$$\frac{1}{u_{eff}(\mathbf{q}=\mathbf{0}, i\nu_n)} + \Pi_{vac}(\mathbf{q}=\mathbf{0}, i\nu_n) \simeq \left[\frac{g^2/(\tilde{\delta} - E_{LP})^2}{[u + g^2/(\tilde{\delta} - E_{LP})]^2} - \frac{M}{4\pi\hbar^2}\frac{1}{E_{LP}}\right](i\nu_n - E_{LP}). \tag{A4}$$

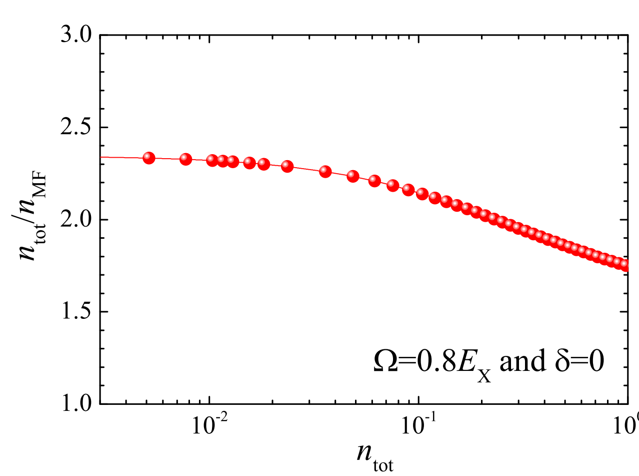

FIG. 8. The ratio n_{tot}/n_{MF} as a function of the total carrier density, at a very strong light-matter coupling $\Omega = 0.8E_X$ and at zero photon detuning $\delta = 0$.

Therefore, we obtain

$$\Gamma_{\rm vac}(\mathbf{q}=\mathbf{0}, i\nu_n) \simeq \frac{4\pi\hbar^2}{M}(-E_{\rm LP})\frac{1}{i\nu_n - E_{\rm LP}} \times \left[1 - \frac{4\pi\hbar^2}{Mu_{\rm LP}^2}\frac{g^2 E_{\rm LP}}{(\tilde{\delta}-E_{\rm LP})^2}\right]^{-1}, \quad (A5)$$

implying

$$C = \frac{4\pi\hbar^2}{M}(-E_{\rm LP}), \quad (A6)$$

$$\xi_{\rm LP}^2(\mathbf{q}=\mathbf{0}) = \left[1 - \frac{4\pi\hbar^2}{Mu_{\rm LP}^2}\frac{g^2 E_{\rm LP}}{(\tilde{\delta}-E_{\rm LP})^2}\right]^{-1}. \quad (A7)$$

APPENDIX B: DENSITY DEPENDENCE OF THE RATIO $n_{\rm tot}/n_{\rm MF}$

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

[1] H. Deng, H. Haug, and Y. Yamamoto, Exciton-polariton Bose-Einstein condensation, Rev. Mod. Phys. **82**, 1489 (2010).

[2] T. Byrnes, N. Y. Kim, and Y. Yamamoto, Exciton-polariton condensates, Nat. Phys. **10**, 803 (2014).

[3] H. Deng, G. Weihs, C. Santori, J. Bloch, and Y. Yamamoto, Condensation of semiconductor microcavity exciton polaritons, Science **298**, 199 (2002).

[4] C. Schneider, K. Winkler, M. D. Fraser, M. Kamp, Y. Yamamoto, E. A. Ostrovskaya, and S. Höling, Exciton-polariton trapping and potential landscape engineering, Rep. Prog. Phys. **80**, 016503 (2017).

[5] M. D. Fraser, S. Höling, and Y. Yamamoto, Physics and applications of exciton-polariton lasers, Nat. Mater. **15**, 1049 (2016).

[6] D. Sanvitto and S. Kna-Cohen, The road towards polaritonic devices, Nat. Mater. **15**, 1061 (2016).

[7] L. Ferrier, E. Wertz, R. Johne, D. D. Solnyshkov, P. Senellart, I. Sagnes, A. Lemare, G. Malpuech, and J. Bloch, Interactions in Confined Polariton Condensates, Phys. Rev. Lett. **106**, 126401 (2011).

[8] A. S. Brichkin, S. I. Novikov, A. V. Larionov, V. D. Kulakovskii, M. M. Glazov, C. Schneider, S. Hling, M. Kamp, and A. Forchel, Effect of Coulomb interaction on exciton-polariton condensates in GaAs pillar microcavities, Phys. Rev. B **84**, 195301 (2011).

[9] S. Kim, B. Zhang, Z. Wang, J. Fischer, S. Brodbeck, M. Kamp, C. Schneider, S. Höfling, and H. Deng, Coherent Polariton Laser, Phys. Rev. X **6**, 011026 (2016).

[10] Y. Sun, P. Wen, Y. Yoon, G. Liu, M. Steger, L. N. Pfeiffer, K. West, D. W. Snoke, and K. A. Nelson, Bose-Einstein Condensation of Long-Lifetime Polaritons in Thermal Equilibrium, Phys. Rev. Lett. **118**, 016602 (2017).

[11] G. Muz-Matutano, A. Wood, M. Johnsson, X. Vidal, B. Q. Baragiola, A. Reinhard, A. Lemare, J. Bloch, A. Amo, G. Nogues, B. Besga, M. Richard, and T. Volz, Emergence of quantum correlations from interacting fibre-cavity polaritons, Nat. Mater. **18**, 213 (2019).

[12] A. Delteil, T. Fink, A. Schade, S. Hling, C. Schneider, and A. İmamoğlu, Towards polariton blockade of confined exciton–polaritons, Nat. Mater. **18**, 219 (2019).

[13] E. Estrecho, T. Gao, N. Bobrovska, D. Comber-Todd, M. D. Fraser, M. Steger, K. West, L. N. Pfeiffer, J. Levinsen, M. M. Parish, T. C. H. Liew, M. Matuszewski, D. W. Snoke, A. G. Truscott, and E. A. Ostrovskaya, Direct measurement of polariton-polariton interaction strength in the Thomas-Fermi regime of exciton-polariton condensation, Phys. Rev. B **100**, 035306 (2019).

[14] J. Hu, Z. Wang, S. Kim, H. Deng, S. Brodbeck, C. Schneider, S. Hling, N. H. Kwong, and R. Binder, Signature of a Bardeen-Cooper-Schrieffer polariton laser, arXiv:1902.00142v1.

[15] C. Ciuti, V. Savona, C. Piermarocchi, A. Quattropani, and P. Schwendimann, Role of the exchange of carriers in elastic exciton-exciton scattering in quantum wells, Phys. Rev. B **58**, 7926 (1998).

[16] F. Tassone and Y. Yamamoto, Exciton-exciton scattering dynamics in a semiconductor microcavity and stimulated scattering into polaritons, Phys. Rev. B **59**, 10830 (1999).

[17] M. M. Glazov, H. Ouerdane, L. Pilozzi, G. Malpuech, A. V. Kavokin, and A. D'Andrea, Polariton-polariton scattering in microcavities: A microscopic theory, Phys. Rev. B **80**, 155306 (2009).

[18] F. Xue, F. Wu, M. Xie, J.-J. Su, and A. H. MacDonald, Microscopic theory of equilibrium polariton condensates, Phys. Rev. B **94**, 235302 (2016).

[19] J. Levinsen, G. Li, and M. M. Parish, Microscopic description of exciton-polaritons in microcavities, Phys. Rev. Research **1**, 033120 (2019).

[20] We note that, although the underlying interaction between electrons and holes is of the Coulomb type, the effective interaction between composite bosons, including both excitons and polaritons, could be described by a short-range Lennard-Jones potential $W[(a_*/r)^{12} - (a_*/r)^6]$, with a strength W and a length scale a_* comparable to the excitonic Bohr radius a_X. At low temperature, only the s-wave channel is important and we then can use an s-wave scattering length a_s to characterize the effective interaction.

[21] M. Schick, Two-dimensional system of hard-core bosons, Phys. Rev. A **3**, 1067 (1971).

[22] C. Mora and Y. Castin, Ground State Energy Of The Two-Dimensional Weakly Interacting Bose Gas: First Correction Beyond Bogoliubov Theory, Phys. Rev. Lett. **102**, 180404 (2009).

[23] In scattering theory, the Born approximation amounts to truncating the Lippmann-Schwinger equation to the first order and taking the bare scattering potential to replace the two-body T-matrix. In the quantum many-body approach, the Born approximation result for the interparticle interaction strength can

often be conveniently obtained, by taking mean-field approximation to the many-body theory. In this context, we take the Born approximation and mean-field approximation as exchangeable terminology.

[24] L. He, H. Lü, G. Cao, H. Hu, and X.-J. Liu, Quantum fluctuations in the BCS-BEC crossover of two-dimensional Fermi gases, Phys. Rev. A **92**, 023620 (2015).

[25] D. S. Petrov, C. Salomon, and G. V. Shlyapnikov, Weakly Bound Dimers of Fermionic Atoms, Phys. Rev. Lett. **93**, 090404 (2004).

[26] I. V. Brodsky, A. V. Klaptsov, M. Yu. Kagan, R. Combescot, and X. Leyronas, Exact diagrammatic approach for dimer-dimer scattering and bound states of three and four resonantly interacting particles, Phys. Rev. A **73**, 032724 (2006).

[27] H. Hu and X.-J. Liu, Quantum fluctuations in a strongly interacting Bardeen-Cooper-Schrieffer polariton condensate at thermal equilibrium, Phys. Rev. A **101**, 011602(R) (2020).

[28] We note that slightly different notations are used in Ref. [27]. There, the Bohr radius and binding energy are denoted by a_B and E_B, respectively. The photon detuning δ is measured from the edge of the band gap E_g and should be identified as $\tilde{\delta}$ in the current paper.

[29] H. Hu, H. Deng, and X.-J. Liu, Two-dimensional exciton-polariton interactions beyond the Born approximation, arXiv:2004.05559.

[30] K. Kamide and T. Ogawa, What Determines the Wave Function of Electron-Hole Pairs in Polariton Condensates?, Phys. Rev. Lett. **105**, 056401 (2010).

[31] T. Byrnes, T. Horikiri, N. Ishida, and Y. Yamamoto, BCS Wave-Function Approach to the BEC-BCS Crossover of Exciton-Polariton Condensates, Phys. Rev. Lett. **105**, 186402 (2010).

[32] M. Yamaguchi, K. Kamide, T. Ogawa, and Y. Yamamoto, BEC–BCS-laser crossover in Coulomb-correlated electron–hole–photon systems, New J. Phys. **14**, 065001 (2012).

[33] L. V. Keldysh, Coulomb interaction in thin semiconductor and semimetal films, JETP Lett. **29**, 658 (1979).

[34] P. Cudazzo, I. V. Tokatly, and A. Rubio, Dielectric screening in two-dimensional insulators: Implications for excitonic and impurity states in graphane, Phys. Rev. B **84**, 085406 (2011).

[35] R. Hanai, P. B. Littlewood, and Y. Ohashi, Photoluminescence and gain/absorption spectra of a driven-dissipative electron-hole-photon condensate, Phys. Rev. B **97**, 245302 (2018).

[36] Y. Ohashi and A. Griffin, BCS-BEC Crossover in a Gas of Fermi Atoms with a Feshbach Resonance, Phys. Rev. Lett. **89**, 130402 (2002).

[37] Y. Ohashi and A. Griffin, Superfluidity and collective modes in a uniform gas of Fermi atoms with a Feshbach resonance, Phys. Rev. A **67**, 063612 (2003).

[38] X.-J. Liu and H. Hu, Self-consistent theory of atomic Fermi gases with a Feshbach resonance at the superfluid transition, Phys. Rev. A **72**, 063613 (2005).

[39] H. Hu, B. C. Mulkerin, U. Toniolo, L. He, and X.-J. Liu, Reduced Quantum Anomaly in a Quasi-Two-Dimensional Fermi Superfluid: Significance of the Confinement-Induced Effective Range of Interactions, Phys. Rev. Lett. **122**, 070401 (2019).

[40] V. Makhalov, K. Martiyanov, and A. Turlapov, Ground-State Pressure of Quasi-2D Fermi and Bose Gases, Phys. Rev. Lett. **112**, 045301 (2014).

[41] M. G. Ries, A. N. Wenz, G. Zn, L. Bayha, I. Boettcher, D. Kedar, P. A. Murthy, N. Neidig, T. Lompe, and S. Jochim, Observation Of Pair Condensation in the Quasi-2D BEC–BCS Crossover, Phys. Rev. Lett. **114**, 230401 (2015).

[42] A. V. Turlapov and M. Y. Kagan, Fermi-to-Bose crossover in a trapped quasi-2D gas of fermionic atoms, J. Phys.: Condens. Matter **29**, 383004 (2017).

[43] J. Levinsen and M. M. Parish, Strongly interacting two-dimensional Fermi gases, in *Annual Review of Cold Atoms and Molecules* (World Scientific, Singapore, 2015), Vol. 3, Chap. 1, pp. 1–75.

[44] B. C. Mulkerin, K. Fenech, P. Dyke, C. J. Vale, X.-J. Liu, and H. Hu, Comparison of strong-coupling theories for a two-dimensional Fermi gas, Phys. Rev. A **92**, 063636 (2015).

[45] H. Hu, X.-J. Liu, and P. D. Drummond, Equation of state of a superfluid Fermi gas in the BCS-BEC crossover, Europhys. Lett. **74**, 574 (2006).

[46] H. Hu, P. D. Drummond, and X.-J. Liu, Universal thermodynamics of strongly interacting Fermi gases, Nat. Phys. **3**, 469 (2007).

[47] R. B. Diener, R. Sensarma, and M. Randeria, Quantum fluctuations in the superfluid state of the BCS-BEC crossover, Phys. Rev. A **77**, 023626 (2008).

[48] J. Keeling, P. R. Eastham, M. H. Szymańska, and P. B. Littlewood, BCS-BEC crossover in a system of microcavity polaritons, Phys. Rev. B **72**, 115320 (2005).

[49] D. S. Petrov, M. A. Baranov, and G. V. Shlyapnikov, Superfluid transition in quasi-two-dimensional Fermi gases, Phys. Rev. A **67**, 031601(R) (2003).

[50] D. S. Citrin and J. B. Khurgin, Microcavity effect on the electron-hole relative motion in semiconductor quantum wells, Phys. Rev. B **68**, 205325 (2003).

[51] O. Bleu, J. Levinsen, and M. M. Parish, Polariton interactions in microcavities with atomically thin semiconductor layers, Phys. Rev. Research **2**, 043185 (2020).

[52] A. E. Golomedov, Yu. E. Lozovik, G. E. Astrakharchik, and J. Boronat, Composite boson description of a low-density gas of excitons, J. Low Temp. Phys. **189**, 300 (2017).

[53] X. Y. Yin, H. Hu, and X.-J. Liu, Cluster Formation in Two-Component Fermi Gases, Phys. Rev. Lett. **123**, 073401 (2019).

[54] X. Y. Yin, H. Hu, and X.-J. Liu, Few-Body Perspective of a Quantum Anomaly in Two-Dimensional Interacting Fermi Gases, Phys. Rev. Lett. **124**, 013401 (2020).