Two-dimensional exciton-polariton interactions beyond the Born approximation

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We provide a many-body theory for the interactions of two-dimensional excitons and polaritons beyond the Born approximation. Taking into account Gaussian quantum fluctuations via the Bogoliubov theory, we find that the two-body interaction strength in two-dimensions has an inverse logarithmic dependence on the scattering length and ground state energy. This leads to a vanishing exciton interaction strength in the zero-momentum limit but a finite polariton interaction strength due to strong light-matter coupling. We also derive the exact Tan relations for exciton-polaritons and calculate Tan’s contact coefficient. We show the polariton interaction strength and Tan’s contact both exhibit an anomalous enhancement at red photon-exciton detuning when the scattering length is large. Our predictions may provide a qualitatively correct guide for studies of exciton and polariton nonlinearities, and suggest a route to achieving strongly nonlinear polariton gases.

Exciton-polaritons are elementary excitations of a semiconductor formed via strong coupling between excitons and photons \[1\]. Due to their half-matter, half-light nature, they form a unique platform for a wide range of novel nonlinear phenomena that are absent in linear optical systems and hard to access in pure matter systems \[2\–7\], ranging from a variety of many-body quantum phases \[8\–10\], resonant parametric scattering \[11\–12\], ultra-low threshold lasing \[13\–14\], to fast and low-power switching \[15\–16\]. With a stronger polariton nonlinearity, polariton blockade \[17\–18\] and all-optical integrated quantum gates \[19\] may also be possible.

While nonlinearity plays a pivotal role in polaritonic phenomena, it has been found to be relatively weak in commonly studied systems, and its origin, controversial. The full solution of the polariton interaction is a formidable quantum mechanics challenge, as we need to solve a six-body problem involves two photons, two electrons and two holes. Instead, most previous studies use the Born approximation, or a mean-field approach \[20\]. The polariton interaction strength \(g_{PP}\) is considered to be directly determined by that of the exciton’s, \(g_{XX}\), as: \(g_{PP} = X_{LP}^2 g_{XX}\), for \(X_{LP}^2\) the Hopfield coefficient, corresponding to the excitation energy in the lower polariton (LP) mode. Treating the exciton scattering in the Born approximation leads to the widely used result \[21\–24\]:

\[
g_{PP}^{(0)} = X_{LP}^4 g_{XX}^{(0)} \approx 6.06 E_X a_X^2, \tag{1}
\]

where \(E_X = \hbar^2/(2m_e a_X^2)\) and \(a_X\) are the binding energy and Bohr radius of excitons with a total mass \(m_X = m_e + m_h\) and a reduced mass \(m_r = m_e m_h/m_X\). However, there is a fundamental conceptual inconsistency. Born approximation, indicated here by the superscript “0”, is often used in three dimensions. But it is known to fail in low dimensions even at the \textit{qualitative} level, due to strong quantum fluctuations \[23\–28\].

In this work, taking into account Gaussian quantum fluctuations in a many-body approach \[23\–28\], we obtain an analytical expression for exciton and polariton interactions in two dimensions (2D) beyond the Born approximation. We show that, while the two-body exciton interaction strength \(g_{XX}\) vanishes in 2D due to quantum fluctuations \[23\–26\], strong coupling with photon introduces a new energy scale and leads to a finite two-body polariton interaction strength \(g_{PP}\) of the form:

\[
g_{PP} = X_{LP}^4 \left( \frac{4 \pi \hbar^2}{m_X} \right) \ln^{-1} \left[ \frac{2}{e^{2 \gamma} m_X a_X^2 |E_{LP}|} \right], \tag{2}
\]

where \(\gamma \approx 0.577\) is Euler’s constant, \(a_s\) is the exciton-exciton s-wave scattering length, and \(E_{LP} = \delta/2 - \sqrt{\delta^2/4 + \Omega^2}\) is the lower polariton energy for photon-exciton detuning \(\delta\) and coupling strength \(\Omega\) \[3\]. We furthermore derive the exact universal Tan relations \[29\–31\] for 2D polaritons and determine Tan contact coefficient \(\mathcal{I}\), which underlies a \(q^{-4}\) tail in the excitonic momentum distribution \(n_X(q \to \infty) \sim \mathcal{I}/q^4\).

Our results reveal that, contrary to the predication Eq. (1), the polariton interaction strength may be greatly enhanced at negative photon-exciton detuning when the exciton scattering length is large, with correspondingly an even more dramatic increase in the Tan contact coefficient \(\mathcal{I}\). These predictions could be experimentally checked in quantum wells \[32\–33\] or van der Waals monolayers \[34\–35\] placed in microcavities. The unusual detuning dependence of the polariton-polariton interaction strength could provide a way to measure the hitherto unknown 2D exciton-exciton scattering lengths \[36\] and to achieve strong polariton nonlinearities in systems with large scattering lengths.

Equation (2) is applicable when excitons can be well regarded as point-like, structureless bosons as in the standard exciton-polariton model, a picture generally adopted by the polariton community \[3\–4\]. We use the zero-temperature Bogoliubov theory \[23\–28\] as a minimal description of those bosons, by assuming all photons and excitons are coherently condensed into the zero-momentum state. The strong Gaussian fluctuations are
then well-characterized by Bogoliubov quasiparticles out of the condensate. In this description, the interaction energy is simply the chemical potential \( \mu \) measured with respect to \( E_{LP} \), i.e., \( E_{\text{int}} = \mu - E_{LP} = g_{PP} n \), which is in turn proportional to the density \( n \) in the dilute limit. We will take advantage of this relation to calculate the two-body polariton-polariton interaction strength \( g_{PP} \), although we solve a many-body problem.

**Model Hamiltonian.** The 2D electron-hole-photon system in microcavities can be described by the following model Hamiltonian \( \mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{LM}} + \mathcal{H}_{\text{int}} \) as

\[
\mathcal{H}_0 = \frac{\hbar^2}{2m_X} \left( \frac{\mathbf{q}^2}{2m_{\text{ph}}} + \delta - \mu \right) \phi_a \phi_a + \xi \phi_a^\dagger \phi_a \mathbf{X}_a \mathbf{X}_a,
\]

\[
\mathcal{H}_{\text{LM}} = \frac{\Omega}{\sqrt{S}} \sum \left[ \phi_a^\dagger \mathbf{X}_a + \phi_a \mathbf{X}_a^\dagger \right],
\]

\[
\mathcal{H}_{\text{int}} = \frac{g}{2S} \sum \mathbf{X}_a^\dagger \mathbf{X}_a, \quad \mathbf{X}_a \equiv \mathbf{X}_{a,\mathbf{q}} \mathbf{X}_{a,\mathbf{q}^\prime}
\]

Here, \( \xi \equiv \hbar^2 q^2/(2m_X) - \mu \) is the excitonic dispersion relation with the chemical potential \( \mu \). \( S \) is the area of the system and hereafter is taken to be unity, \( \phi_a \) and \( \mathbf{X}_a \) are the annihilation field operators for photons and excitons, respectively. The mass of cavity photons \( m_{\text{ph}} \) is typically several orders smaller than the exciton mass \( m_X \). In the interaction Hamiltonian \( \mathcal{H}_{\text{int}} \), \( g \) is a bare exciton interaction strength, which is to be replaced by the exciton-exciton s-wave scattering length \( a_s \) according to [22, 23],

\[
\frac{1}{g} + \sum_q \left( \frac{\hbar^2 q^2}{m_X} + \varepsilon_c \right)^{-1} = \frac{m_X}{4 \pi \hbar^2} \ln \left( \frac{4 \pi \hbar^2}{m_X a_s^2 \varepsilon_c} \right).
\]

Here \( \varepsilon_c > 0 \) is an arbitrary energy used to regularize the infrared divergence, which is unavoidable in 2D [23, 24].

In the absence of the photon field, the model Hamiltonian describes a weakly interacting 2D Bose gas and has been solved by Popov [24], based on whose work the density equation of state within the Bogoliubov approximation was obtained as [26, 23, 30]:

\[
n(\mu_B) = \frac{m_X \mu_B}{4 \pi \hbar^2} \ln \left( \frac{4 \pi \hbar^2}{m_X \mu_B a_s^2} \right).
\]

This implies that the effective interaction strength \( g_{XX} = \mu_B/n \) depends logarithmically on the chemical potential \( \mu_B \) or the density \( n \), and consequently vanishes identically in the dilute limit (i.e., \( n \to 0 \)). When we add the photon field, coherent superposition of photons and excitons gives rise to two polariton branches in the energy spectrum [3]. Focusing on LP only, the creation field operator can be written as \( P^\dagger \equiv \sqrt{1 - X_{LP}^\dagger X_{LP}} \). As there is no interaction between photons, the interaction between polaritons should come from the excitonic part. By rewriting the interaction Hamiltonian \( \mathcal{H}_{\text{int}} \) in terms of \( P \) and \( P^\dagger \), we then have the naive expression \( g_{PP} \approx X_{LP}^4 g_{XX} \), as we already see in Eq. [11] within the Born approximation. Beyond the Born approximation, \( g_{PP} \) therefore should vanish in the dilute limit, exactly in the same way as the effective exciton interaction strength \( g_{XX} \). This disagrees with experimental findings [32, 33]. To solve this apparent contradiction, we note that there could be virtual excitations from the LP branch to the upper-polariton branch, by the residual scattering terms (generated when we rewrite \( \mathcal{H}_{\text{int}} \) in terms of \( P \) and \( P^\dagger \)). These virtual excitations may render the polariton-polariton interaction strength finite as we show below using the Bogoliubov theory.

**Bogoliubov theory.** At zero temperature \( T = 0 \), both photons and excitons macroscopically condense into zero-momentum states with wave-functions \( \phi_0 \) and \( X_0 \), respectively. To the leading order, the mean-field thermal potential takes the form,

\[
\Omega_0(\mu) = (\delta - \mu) \phi_0^2 + 2\Omega \phi_0 X_0 - \mu X_0^2 + \frac{g}{2} X_0^4.
\]

By minimizing \( \Omega_0(\mu) \), we obtain \( gX_0^2 = \mu + \Omega^2/(\delta - \mu) \) and \( \Omega(\mu) = -\mu + \Omega^2/(\delta - \mu)^2/2g \).

To take into account crucial quantum fluctuations in 2D, we rewrite the Hamiltonian in terms of \( \delta \phi = \phi - \phi_0 \) and \( \delta X = X - X_0 \) and keep only the bilinear terms at the Gaussian level. We then obtain the inverse Green function \( \mathcal{G}^{-1}(q, \nu_n) \) of the Bogoliubov quasiparticles [23],

\[
-\mathcal{G}^{-1} = \begin{pmatrix}
-n\nu + A_q & 0 & \Omega & 0 \\
0 & -n\nu + A_q & 0 & \Omega \\
\Omega & 0 & -n\nu + B_q & C \\
0 & \Omega & C & -n\nu + B_q
\end{pmatrix}
\]

where \( \nu_n \equiv 2\pi nk_BT \) \((n \in \mathbb{Z})\) are bosonic Matsubara frequencies and we have introduced the notations,

\[
A_q = \frac{\hbar^2 q^2}{2m_{\text{ph}}} + \delta - \mu,
\]

\[
B_q = \frac{\hbar^2 q^2}{2m_X} - \mu + 2gX_0^2 - \frac{\hbar^2 q^2}{2m_X} + \mu + \frac{2\Omega^2}{\delta - \mu},
\]

\[
C = gX_0^2 + \mu + \frac{\Omega^2}{\delta - \mu}.
\]

As the lowest attainable chemical potential is \( E_{LP} \), i.e., \( \mu = E_{LP} + \mu_B \) where \( \mu_B > 0 \) in the dilute limit we find \( C \approx [1 + \Omega^2/(\delta - E_{LP})^2]^{1/2} = X_{LP}^2 \mu_B > 0 \). By solving \( \det \left[ -\mathcal{G}^{-1}(q, \nu_n \rightarrow E) \right] = 0 \), we obtain the quasiparticle energy spectrum,

\[
E_{q \pm} = K_{q \pm} + \Omega^2 + \frac{K_{q \pm}^2}{2} \sqrt{\left( A_q + B_q \right)^2 - C^2} \Omega^2,
\]

where we have defined \( K_{q \pm} = [A_q^2 + B_q^2 + C^2]/2 \).

At the Gaussian level for quantum fluctuations, quasiparticles are approximately treated as non-interacting.
particles. Thus, their contribution to the thermodynamic potential can be written down straightforwardly [37],

\[
\delta \Omega_g = \frac{k_B T}{2} \sum_{\mathbf{q}, \nu_n} \ln \det [-\Pi^{-1}(\mathbf{q}, i\nu_n)] e^{i\nu_n \phi},
\]

(12)

where the convergence factor \(e^{i\nu_n \phi}\) is used to regularize the divergence at \(\nu_n \to \pm \infty\). As we discuss in detail in Supplemental Material [36], the summation over the bosonic Matsubara frequencies can be explicitly performed and at zero temperature we find \(\delta \Omega_g^{(T=0)} = \sum_{\mathbf{q}} [E_{\mathbf{q}+} + E_{\mathbf{q}-} - A_q - B_q]/2\), which formally diverges. However, this ultraviolet divergence can be exactly cancelled by the same divergence in the mean-field thermodynamic potential \(\Omega_0\). By putting these two contributions together, i.e., \(\Omega = \Omega_0 + \delta \Omega_g^{(T=0)}\), we arrive at [36]

\[
\Omega = \frac{m_X C^2}{8 \pi \hbar^2} \ln \left[ \frac{4}{\epsilon^{2\gamma}} \frac{\hbar^2}{m_X a^2 \epsilon} \right] + \frac{1}{2} \sum_{\mathbf{q}} [E_{\mathbf{q}+} + E_{\mathbf{q}-} - A_q - B_q + C^2 / \hbar^2 q^2 / m_X + \epsilon].
\]

(13)

At nonzero light-matter coupling, interestingly, the integration over the momentum in the above can be worked out analytically in the infinite mass ratio limit \(m_X/m_{ph} \to \infty\). We find that [36],

\[
\Omega = \frac{m_X}{8 \pi \hbar^2} \left( \mu + \frac{\Omega^2}{\delta - \mu} \right)^2 \ln \left[ \frac{2 \hbar^2 (\delta - \mu)}{e^{2\gamma} m_X a^2 \Omega^2} \right].
\]

(14)

By keeping the leading term in powers of \(\mu_B = \mu - E_{LP}\) and taking derivative of \(\Omega\) with respect to \(\mu_B\), i.e., \(n = -\partial \Omega / \partial \mu_B\), we obtain

\[
n = \frac{\mu_B}{X_{LP}^2} \left( \frac{m_X}{4 \pi \hbar^2} \right) \ln \left[ \frac{2 \hbar^2}{e^{2\gamma} m_X a^2 |E_{LP}|} \right].
\]

(15)

and hence the polariton-polariton interaction strength in Eq. (4). By comparing the above density equation with Eq. (7), we see that the small chemical potential \(\mu_B\) in the logarithm is now replaced with a characteristic finite LP energy, due to the virtual scatterings between the two polariton branches. As a result, the polariton-polariton interaction strength in Eq. (2) becomes finite in the dilute limit. This observation is the first main result of our work. It is also applicable to the case of \(N\) quantum wells, where the polariton interaction is reduced by a factor of \(N\). [36]

Validity of our results. Eq. (2) is an exact two-body result, valid as long as the bosonic model holds. This implies that we need \(\Omega < \Omega_c \ll E_X\) and the density \(n < n_c \sim 0.01 a_X^2\), so that the internal fermionic degrees of freedom of excitons are frozen and do not lead to observable effects. To estimate \(\Omega_c\), we compare our results with a fermionic toy model without such a restriction, where the Coulomb interaction is approximately replaced by a contact interaction and electrons and holes are assumed to have the same mass \(m_e = m_h = M\). It can be reliably solved by using a fermionic Gaussian pair fluctuation (GPF) theory, which in the dilute limit recovers the bosonic Bogoliubov theory [38]. Within the toy model, the exciton \(s\)-wave scattering length \(a_s \sim 1.12e^{-\gamma} a_X\) is known [27]. Therefore, we can compare the predictions from both the bosonic and fermionic models under the same condition. As shown in Fig. 1 we find a good agreement at \(\Omega = 0.1 E_X\), indicating \(\Omega_c \sim 0.1 E_X\).

Experimentally, the polariton interaction has been reported for MoSe\(_2\) monolayers at \(\Omega = 5.0\) meV [34] or \(\Omega = 17.2\) meV [33] near zero detuning. These light-matter couplings are much smaller than the exciton binding energy \(E_X \sim 500\) meV [40]. Using Eq. (2) and \(m_e \simeq m_h \simeq 0.5m_0\) for MoSe\(_2\), we obtain \(g_{PP} \sim 0.1\) meV \(\cdot\) \(\mu\)m\(_2\), which is consistent with the experimental data \(g_{PP} = 0.01 - 1.0\) meV \(\cdot\) \(\mu\)m\(_2\) [34] [32].

Anomalous interaction enhancement. The inverse logarithmic dependence of the polariton-polariton interaction strength \(g_{PP}\) on the LP energy \(E_{LP}\) shown in Eq. (2) is nontrivial. As \(|E_{LP}|\) can be enlarged by tuning the photon detuning even at \(\Omega < \Omega_c\), we find the second main result of our work that the polariton interaction could be anomalously enhanced at a large red detuning. To see this, for the Coulomb interaction let us recast the expression of \(g_{PP}\) into the form,

\[
g_{PP} \sim \frac{g_{PP}^{(0)}}{g_{XX}^{(0)}} \left( \frac{X_{LP}^2}{3.03 m_X^2} \left[ -\ln \left| E_{LP} / E_X \right| + C_{PP} \right] \right),
\]

(16)

where \(C_{PP} \equiv 2\ln(a_s/a_X) + \ln(m_e/m_X) + 2\gamma - 2\ln 2\). Clearly, a resonance appears at \(|E_{LP}| = C_{PP}/E_X\), when the photon field is significantly occupied and the scat-
tering between excitons is then drastically altered. Our perturbative Bogoliubov theory breaks down at resonance. However, away from the resonance the qualitative anomalous enhancement seems to be physical.

In Fig. 2 we report the polariton-polariton interaction strengths for GaAs quantum well (a) and TMD monolayer (b) in microcavities, with masses $m_c \simeq 0.067 m_0$ and $m_h \simeq 0.45 m_0$ (GaAs) [3] and $m_e \simeq m_h \simeq 0.5 m_0$ (TMD) [40], respectively. As the exciton-exciton $s$-wave scattering length $a_s$ remains elusive for the Coulomb interaction in 2D [30] and might be tunable [30, 41], we consider three likely choices, as inspired by the result in three dimensions (i.e., $a_s \sim a_X$) [42]. In comparison to the Born approximation result $g_{PP}^{(0)}$, as shown in Fig. 2(a), we find the ratio $g_{PP}/g_{PP}^{(0)}$ decreases monotonically with increasing photon detuning. In contrast, measured in units of $g_{XX}^{(0)}$ as plotted in Fig. 2(b), the anomalous enhancement becomes less apparent, except at large $a_s \sim 1.5 a_X$ where the rise at red detuning is always significant. This sensitive dependence of the polariton interaction on $a_s$ provides a unique way to measure the long-sought exciton-exciton scattering length in 2D semiconductor materials [30].

**Tan relations.** We now consider the universal relations which govern the short-range, large-momentum and high-energy behaviors of a quantum many-body system [29–31]. In these exact relations, the central role is played by Tan’s contact coefficient $I = \langle m_X q^2/\hbar^2 \rangle \int d\mathbf{r} \langle \mathcal{X}^\dagger(\mathbf{r})\mathcal{X}(\mathbf{r})\mathcal{X}(\mathbf{r})\rangle$. As discussed in detail in Supplemental Material, we derive the adiabatic and energy relations [30],

\[
\left[ \frac{\partial \Omega}{\partial \ln a_s} \right]_{\mu,S} = \frac{\hbar^2}{4\pi m_X} I, \\
\mathcal{F}_X + \mathcal{E}_{\text{int}} = \sum_q \frac{\hbar^2 q^2}{2 m_X} \tilde{n}_X(q) + \frac{(\ln 2 - \gamma) h^2 I}{4 \pi m_X},
\]

where $\tilde{n}_X(q) \equiv n_X(q) - I/[q^2(q^2 + a_s^{-2})]$, $\mathcal{F}_X$ and $\mathcal{E}_{\text{int}}$ are the excitonic kinetic energy and interaction energy, respectively. By applying the adiabatic relation to Eq. (14), we obtain within the Bogoliubov approximation,

\[
I = \frac{m_X^2 C^2}{\hbar^4} \simeq \frac{(16\pi^2 n^2) X_{LP}^4}{\ln^2 [e^{2\gamma} m_X a_s^2 |E_{LP}|/(2\hbar^2)]}.
\]

Figure 3 presents the detuning dependence of the contact coefficient for GaAs quantum well (dashed line) and TMD monolayer (solid line) at $a_s = a_X$. In accordance with the anomalous enhancement in the polariton interaction, we also observe a dramatic increase in the contact coefficient at red detuning, which can be measured from the universal $q^{-4}$ tail in the excitonic momentum distribution $n_X(q)$.

**Conclusions.** We have derived an analytic expression for interactions of two-dimensional exciton-polaritons. Compared to the previous constant two-body interaction strength derived within the Born approximation, our result shows a logarithmic dependence on both the exciton
s-wave scattering length and shift of the polariton energy from the bare exciton energy. Such a dependence leads to a counter intuitive, large enhancement in the polariton-polariton interaction strength, and Tan’s contact coefficient at red photon-exciton detunings when the scattering length is greater than the exciton Bohr radius. Therefore our result suggests a way to measure the 2D exciton scattering length and reveals the possibility of achieving a strongly nonlinear polariton gas in materials with a large exciton scattering length.

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Appendix A: Quantum fluctuation thermodynamic potential

At the Gaussian level for quantum fluctuations, Bogoliubov quasiparticles are treated as non-interacting and described by the Green function,

\[ -\mathcal{G}^{-1}(q, i\nu_n) = \begin{pmatrix}
-n\nu_n + A_q & 0 & \Omega & 0 \\
0 & i\nu_n + A_q & 0 & \Omega \\
\Omega & 0 & -i\nu_n + B_q & C \\
0 & \Omega & C & i\nu_n + B_q
\end{pmatrix}, \]

(A1)

where \( \nu_n = 2\pi nk_B T \) \((n \in \mathbb{Z})\) are bosonic Matsubara frequencies, and

\[ A_q \equiv \frac{\hbar^2 q^2}{2m_{ph}} + \delta - \mu, \]

(A2)

\[ B_q \equiv \frac{\hbar^2 q^2}{2m_X} - \mu + 2gX_0^2 = \frac{\hbar^2 q^2}{2m_X} + \mu + \frac{2\Omega^2}{\delta - \mu}, \]

(A3)

\[ C \equiv gX_0^2 = \mu + \frac{\Omega^2}{\delta - \mu}. \]

(A4)

We note that the chemical potential satisfies

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

(A5)

As a result, we have \( A_q > 0, B_q > 0 \) and \( C > 0 \). In particular, by writing \( \mu = E_{LP} + \mu_B \) with \( \mu_B > 0 \), we find

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

(A6)

See Supplemental Material for the detailed information on the determination of the quantum fluctuation thermodynamic potential, the tunability of the exciton-exciton \( s \)-wave scattering length, the derivation of the exact universal Tan relation, the direct calculation of the photonic and excitonic momentum distributions within the Bogoliubov theory, and the results for multiple quantum wells.

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in the dilute zero-density limit (i.e., $\mu_B \to 0$). The poles of the Green function give the energy spectrum of Bogoliubov quasiparticles. We therefore solve the eigenvalue equation,

$$\det \left[ \mathcal{D}^{-1}(\mathbf{q}, \nu_n) - E \right] = E^4 - (A_q^2 + B_q^2 - C^2 + 2\Omega^2)E^2 + \left( A_q^2 B_q^2 - A_q^2 C^2 - 2A_q B_q \Omega^2 + \Omega^4 \right) = 0,$$

and find the quasiparticle energy spectrum,

$$E_{q\pm}^2 = \left( \frac{A_q^2 + B_q^2 - C^2}{2} + \Omega^2 \right) \pm \sqrt{\left( \frac{A_q^2 - B_q^2 + C^2}{2} \right)^2 + \left[ (A_q + B_q)^2 - C^2 \right] \Omega^2}.$$  \hspace{1cm} (A8)

It is easy to check that at zero momentum $q = 0$, the lower spectrum $E_{q-} = 0$. This is anticipated, as the quasiparticle spectrum must have a gapless Goldstone model, as a result of the $U(1)$ symmetry breaking.

For non-interacting bosons, their thermodynamic potential takes the form [37],

$$\delta \Omega_q = \frac{k_B T}{2} \sum_{\mathbf{q}, \nu_n} \ln \det \left[ -\mathcal{D}^{-1}(\mathbf{q}, \nu_n) \right] e^{i\nu_n 0^+} = \frac{k_B T}{2} \sum_{\mathbf{q}, \nu_n} \ln \left[ (\nu_n^2 + E_{q+}^2) (\nu_n^2 + E_{q-}^2) \right] e^{i\nu_n 0^+}. \hspace{1cm} (A9)$$

Here, it is necessary to add the convergence factor $e^{i\nu_n 0^+}$ to regularize the ultraviolet divergence at $\nu_n \to \pm \infty$. This is required even for the simplest case of single-component non-interacting bosons with dispersion relation $\xi_q = \hbar^2 q^2/(2M) - \mu > 0$, where the thermodynamic potential is known as,

$$\Omega_B = \frac{k_B T}{2} \sum_{\mathbf{q}, \nu_n} \ln \left[ (\nu_n^2 + \xi_q^2) e^{i\nu_n 0^+} = \frac{k_B T}{2} \sum_{\mathbf{q}, \nu_n} \ln [i\nu_n - \xi_q] e^{i\nu_n 0^+} = \frac{1}{\exp[\xi_q/(k_B T)] - 1} \int_{T = 0} \frac{d\nu}{\nu} \right] (A10)$$

Let us now subtract this zero contribution, i.e.,

$$\frac{k_B T}{2} \sum_{\mathbf{q}, \nu_n} \ln \left[ (\nu_n^2 + A_q^2) (\nu_n^2 + B_q^2) \right] e^{i\nu_n 0^+} = 0 \hspace{1cm} (A11)$$

from the thermodynamic potential $\delta \Omega_g^{(T=0)}$. We obtain,

$$\delta \Omega_g^{(T=0)} = \frac{k_B T}{2} \sum_{\mathbf{q}, \nu_n} \ln \left[ (\nu_n^2 + E_{q+}^2) (\nu_n^2 + E_{q-}^2) \right] \left[ (\nu_n^2 + A_q^2) (\nu_n^2 + B_q^2) \right], \hspace{1cm} (A12)$$

where the convergence factor has been removed, as the integrand now vanishes in the limit $\nu_n \to \pm \infty$ and the integral converges. At zero temperature, by using the identity (i.e., $\nu_n \to \omega$)

$$k_B T \sum_{\nu_n} \ln \left[ \frac{\nu_n^2 + E^2}{\nu_n^2 + \xi^2} \right] = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \frac{\omega^2 + E^2}{\omega^2 + \xi^2} = E - \xi, \hspace{1cm} (A13)$$

we obtain,

$$\delta \Omega_g^{(T=0)} = \frac{1}{2} \sum_q \left[ E_{q+} + E_{q-} - A_q - B_q \right]. \hspace{1cm} (A14)$$

It is worth noting that the integrand in $\delta \Omega_g^{(T=0)}$ is formally divergent. To see this, let us simply consider a zero light-matter coupling $\Omega = 0$, so the photon field is decoupled from the exciton field. In this case, we find that $C = \mu > 0$, $B_q = \hbar^2 q^2/(2m_X) + \mu$, and

$$E_{q+} = A_q, \hspace{1cm} (A15)$$

$$E_{q-} = \sqrt{\frac{\hbar^2 q^2}{2m_X} + 2\mu} \left( \frac{\hbar^2 q^2}{2m_X} + 2\mu \right) \sqrt{\frac{\hbar^2 q^2}{2m_X} + 2\mu}. \hspace{1cm} (A16)$$

Therefore, at large momentum the integrand will be

$$E_{q-} - B_q = \sqrt{\frac{\hbar^2 q^2}{2m_X} \left( \frac{\hbar^2 q^2}{2m_X} + 2\mu \right) - \left( \frac{\hbar^2 q^2}{2m_X} + \mu \right) \sim - \frac{1}{2} \frac{\mu^2}{\hbar^2 q^2/(2m_X) + \mu}. \hspace{1cm} (A17)$$
It is then easy to check the integral of $\delta\Omega_q^{(T=0)}$ is logarithmically divergent. This divergence is actually anticipated, as the mean-field Gross-Pitaevskii thermodynamic potential

$$\Omega_0 = -\frac{[\mu + \Omega^2/(\delta - \mu)]^2}{2g}$$

(A18)

is equally logarithmically divergent. These two divergences will be exactly cancelled once we add the two thermodynamic potentials together, i.e., $\Omega = \Omega_0 + \delta\Omega_q^{(T=0)}$. By expressing the bare interaction strength $g$ in terms of the exciton-exciton $s$-wave scattering length \[28\], i.e.,

$$\frac{1}{g} = \frac{m_X}{4\pi \hbar^2} \ln \left[ \frac{4}{e^{2\gamma} m_X a_s^2 \varepsilon_c} \right] - \sum_q \left[ \frac{\hbar^2 q^2}{m_X} + \varepsilon_c \right]^{-1},$$

(A19)

we arrive at,

$$\Omega = -\frac{m_X C^2}{8\pi \hbar^2} \ln \left[ \frac{4}{e^{2\gamma} m_X a_s^2 \varepsilon_c} \right] + \frac{1}{2} \sum_q \left[ E_{q+} + E_{q-} - A_q - B_q + \frac{C^2}{\hbar^2 q^2/m_X + \varepsilon_c} \right].$$

(A20)

1. Density equation of state of excitons

To obtain an analytic expression for the thermodynamic potential $\Omega$, let us first check the case of excitons in the absence of the light-matter coupling, $\Omega = 0$. As mentioned earlier, we would have $C = \mu = \mu_B > 0$. By choosing a cut-off energy $\varepsilon_c = \mu_B$, we find that the integral in $\Omega$ is,

$$\sum_q \left[ E_{q+} + E_{q-} - A_q - B_q + \frac{C^2}{\hbar^2 q^2/m_X + \varepsilon_c} \right] = \sum_q \left[ \frac{\hbar^2 q^2}{2m_X} \left( \frac{\hbar^2 q^2}{2m_X} + 2\mu_B \right) - \left( \frac{\hbar^2 q^2}{2m_X} + \mu_B \right) + \frac{\mu_B^2}{\hbar^2 q^2/m_X + \mu_B} \right],$$

(A21)

$$\equiv \frac{m_X \mu_B^2}{8\pi \hbar^2} \int_0^\infty dx \left[ \sqrt{x (x + 2)} - (x + 1) + \frac{1}{2x + 1} \right],$$

(A22)

$$\equiv \frac{m_X \mu_B^2}{8\pi \hbar^2} \frac{1}{2},$$

(A23)

where in the second equation, we have introduced a dimensionless variable $x \equiv \hbar^2 q^2/(2m_X \mu_B)$. Therefore, we obtain the thermodynamic potential \[29\],

$$\Omega = -\frac{m_X \mu_B^2}{8\pi \hbar^2} \ln \left[ \frac{4}{e^{2\gamma} m_X a_s^2 \mu_B} \right] + \frac{1}{2} \left( \frac{m_X}{8\pi \hbar^2} \right) \mu_B^2 = -\frac{m_X \mu_B^2}{8\pi \hbar^2} \ln \left[ \frac{4 \mu_B}{e^{2\gamma + 1/2} m_X a_s^2 \mu_B} \right].$$

(A24)

By taking the derivative with respect to the chemical potential $\mu_B$, we obtain the density equation of state for an excitonic gas,

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

(A25)

which is already shown in the main text.

2. Density equation of state of polaritons

Let us now consider the thermodynamic potential in the presence of the light-matter coupling $\Omega \neq 0$ and in the limit of an infinitely large mass ratio $m_X/m_{ph} \rightarrow \infty$. In this limit, $A_q$ is (infinitely) large for any nonzero momentum. Therefore, we may approximate,

$$E_{q\pm}^2 \simeq \left( \frac{A_q^2 + B_q^2 - C^2}{2} + \Omega^2 \right) \pm \left( \frac{A_q^2 - B_q^2 + C^2}{2} \right) \left\{ 1 + \frac{2\Omega^2 (A_q + B_q)^2 - C^2}{(A_q^2 - B_q^2 + C^2)^2} \right\}.$$

(A26)
It is then easy to check that,

\[ E_{q+}^2 \simeq A_q^2 + \frac{2A_q (A_q + B_q)}{A_q^2 - B_q^2 + C^2} \Omega^2, \]

(A27)

\[ E_{q-}^2 \simeq (B_q^2 - C^2) - \frac{2 [(A_q + B_q) B_q - C^2]}{A_q^2 - B_q^2 + C^2} \Omega^2. \]

(A28)

In the limit of \( m_X/m_{\text{ph}} \gg 1 \), we may neglect the second terms in \( E_{q+}^2 \). In other words, the dispersion relations of the photon field and exciton field are effectively decoupled, although the excitonic dispersion is still strongly affected by the light-matter coupling. Therefore, we find that,

\[ \sum_q \left[ E_{q+} + E_{q-} - A_q - B_q + \frac{C^2}{\hbar^2 q^2/m_X + \epsilon_c} \right] \simeq \sum_q \left[ \sqrt{B_q^2 - C^2} - B_q + \frac{C^2}{2B_q} \right] + \sum_q \left[ \frac{C^2}{\hbar^2 q^2/m_X + \epsilon_c} - \frac{C^2}{2B_q} \right]. \]

(A29)

The first integral can be casted into the form (i.e., \( y = B_q/C - 1 \)),

\[ \sum_q \left[ \sqrt{B_q^2 - C^2} - B_q + \frac{C^2}{2B_q} \right] = \frac{m_X C^2}{4\pi \hbar^2} \tilde{I}_1, \]

(A30)

where the dimensionless integral \( \tilde{I}_1 \) is

\[ \tilde{I}_1 = 2 \int_{B_0}^{\infty} dy \left[ \sqrt{y (y + 2)} - (y + 1) + \frac{1}{2y + 2} \right] \]

(A31)

and \( \tilde{B}_0 \equiv B_q=0/C = 1 + \Omega^2/|\langle \delta - \mu \rangle C | \geq 1 \). Actually, with a nonzero light-matter coupling \( \Omega \neq 0 \), \( \tilde{B}_0 \to +\infty \) in the dilute limit since \( C \to 0^+ \). It is easy to check that,

\[ \tilde{I}_1 = \left[ (y + 1) \sqrt{y (y + 2)} - 2 \text{arcsinh} \sqrt{\frac{y}{2} - y^2 - 2y + \ln (2y + 2)} \right]_{\tilde{B}_0 - 1}^{\infty}, \]

(A32)

\[ = \left( \tilde{B}_0^2 - \frac{1}{2} - \tilde{B}_0 \sqrt{\tilde{B}_0^2 - 1} \right) + 2 \text{arcsinh} \sqrt{\frac{\tilde{B}_0 - 1}{2} - \ln \left( 2\tilde{B}_0 \right)}. \]

(A33)

As \( \tilde{B}_0 \to +\infty \), we find that

\[ \tilde{I}_1 \simeq \ln \left( \frac{\tilde{B}_0 - 1}{\tilde{B}_0} \right) \simeq 0. \]

(A34)

On the other hand, the second integral take the form,

\[ \sum_q \left[ \frac{C^2}{\hbar^2 q^2/m_X + \epsilon_c} - \frac{C^2}{\hbar^2 q^2/m_X + 2C + 2\Omega^2/\langle \delta - \mu \rangle} \right] = \frac{m_X C^2}{4\pi \hbar^2} \tilde{I}_2 \]

(A35)

where the dimensionless integral \( \tilde{I}_2 \) is

\[ \tilde{I}_2 = \ln \left( \frac{2\tilde{B}_0}{\epsilon_c/C} \right). \]

(A36)

Therefore, the dimensionless integral \( \tilde{I} = \tilde{I}_1 + \tilde{I}_2 \) is

\[ \tilde{I} = \ln \left( \frac{\tilde{B}_0 - 1}{\tilde{B}_0} \right) + \ln \left( \frac{2\tilde{B}_0}{\epsilon_c/C} \right) = \ln \left[ \frac{2\Omega^2/\langle \delta - \mu \rangle}{\epsilon_c} \right], \]

(A37)

and we obtain that

\[ \sum_q \left[ E_{q+} + E_{q-} - A_q - B_q + \frac{C^2}{\hbar^2 q^2/m_X + \epsilon_c} \right] = \frac{m_X C^2}{4\pi \hbar^2} \ln \left[ \frac{2\Omega^2/\langle \delta - \mu \rangle}{\epsilon_c} \right]. \]

(A38)
We note that, the above integral has also been numerically evaluated (in suitable dimensionless form) for a given mass ratio \( m_X/m_{ph} \). We find that our analytic expression in Eq. (A38) is essentially exact for a realistic mass ratio \( m_X/m_{ph} \sim 10^4 \). We note also that, if the light-matter coupling \( \Omega = 0 \), we would have \( \tilde{B}_0 = 1 \). The dimensionless integrals are then \( \tilde{I}_1 = 1/2 - \ln 2 \) and \( \tilde{I}_2 = \ln[2 \mu_B/\epsilon_c] \), respectively. Therefore, we find that \( \tilde{I} = 1/2 + \ln[\mu_B/\epsilon_c] \), which is 1/2 if we take \( \epsilon_c = \mu_B \). We then recover Eq. (A23), as one may anticipate.

By substituting Eq. (A38) into Eq. (A20), we finally obtain,

\[
\Omega = -\frac{m_X}{8\pi\hbar^2} \left[ \mu + \frac{\Omega^2}{\delta - \mu} \right]^2 \ln \left[ \frac{2}{e^{2\gamma} m_X a_s^2 \Omega^2} \right].
\]  

(A39)

By expanding \( \mu = E_{LP} + \mu_B \), for small \( \mu_B \), we have,

\[
\mu + \frac{\Omega^2}{\delta - \mu} \simeq \frac{\mu_B}{X_{LP}^2},
\]

(A40)

\[
\frac{\Omega^2}{\delta - \mu} \simeq |E_{LP}|.
\]

(A41)

Therefore, we arrive at,

\[
\Omega = -\frac{\mu_B^2}{X_{LP}^4} \left( \frac{m_X}{8\pi\hbar^2} \right) \ln \left[ \frac{2}{e^{2\gamma} m_X a_s^2 |E_{LP}|} \right].
\]

(A42)

Appendix B: Tunability of the exciton-exciton s-wave scattering length

Although the underlying interaction between electrons and holes in semiconductor quantum wells or atomically thin transition-metal-dichalcogenides (TMD) monolayers is of the Coulomb type, the effective interaction between composite excitons \( V_{XX}(r) \) could be described by a short-range Lennard-Jones potential, i.e.,

\[
V_{XX} \simeq W \left[ \left( \frac{a_s}{r} \right)^{12} - \left( \frac{a_s}{r} \right)^{6} \right],
\]

(B1)

with a strength \( W \) and a length scale \( a_s \) comparable to the excitonic Bohr radius \( a_X \). At low temperature, only the \( s \)-wave channel is important and we then can use a single \( s \)-wave scattering length \( a_s \) to characterize the effective interaction. This was illustrated by a recent Monte-Carlo simulation in three dimensions with the \( 1/r \) Coulomb interaction [42]. It was found that the exciton-exciton \( s \)-wave scattering length is comparable to the exciton Bohr radius, \( a_s \sim a_X \). An exact solution for the four-body problem with long-range interaction such as the Coulomb interaction is extremely difficult and is not available.

In real materials, the Coulomb-like interactions among electrons and holes take the following screened potential form [41],

\[
V_C^{\sigma\sigma'}(r) = \chi_{\sigma\sigma'} \frac{\pi e^2}{2\epsilon_0 r_0} \left[ H_0 \left( \frac{r}{r_0} \right) - Y_0 \left( \frac{r}{r_0} \right) \right],
\]

(B2)

where \( \chi_{\sigma\sigma'} = +1 \) for \( \sigma = \sigma' \) and \( \chi_{\sigma\sigma'} = -1 \) for \( \sigma \neq \sigma' \), and the spin index \( \sigma \) stands for either electrons or holes, \( \epsilon_0 \) is the dielectric constant of the substrate surrounding the quantum well or TMD monolayer, \( H_0(x) \) 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 between the quantum well or TMD monolayer and the substrate, which strongly modifies the Coulomb interaction at short distance [41]. As a result, the exciton-exciton \( s \)-wave scattering length \( a_s \) could depend on the effective screening length \( r_0 \) and the dielectric constant \( \epsilon_0 \). Therefore, by carefully designing/choosing the materials, we may have the ability to tune the exciton-exciton \( s \)-wave scattering length \( a_s \).

Appendix C: Universal Tan relations

In 2005, Shina Tan derived a set of exact universal relations to describe the short-range, large-momentum and high-energy behaviors of a quantum many-body system interacting via a short-range potential [29 31]. These relations
are linked by Tan’s contact coefficient \( I \). In ultracold atomic physics, the universal Tan relations help a lot for us to understand the fundamental interacting Fermi gases and Bose gases. Here, we generalize Tan relations to the exciton-polariton system, following the work by Braaten and Platter [31].

For exciton-polaritons, the contact coefficient can be formally defined by,

\[
I = \frac{m_X^2 g^2}{\hbar^4} \int dr \langle X^\dagger(r)X^\dagger(r)X(r)X(r) \rangle,
\]

(C1)

where the average \( \langle ... \rangle \) is taken for any quantum states. It is worth noting that the bare exciton-exciton interaction strength \( g \) is vanishingly small in the sense of its regularization, see Eq. (A19). However, this smallness will be compensated by the divergence in \( \langle X^\dagger X^\dagger XX \rangle \), resulting in a finite contact coefficient. To see this, let us recall that

\[
\frac{\partial g}{\partial \ln a_s} = -g^2 \left( \frac{\partial g^{-1}}{\partial \ln a_s} \right) = \frac{m_X}{2\pi \hbar^2 g^2},
\]

(C2)

and apply the Hellmann–Feynman theorem to the total energy of the system,

\[
\left( \frac{\partial E}{\partial \ln a_s} \right)_{S,N} = \frac{1}{2} \left( \frac{\partial g}{\partial \ln a_s} \right) \int dr \langle X^\dagger(r)X^\dagger(r)X(r)X(r) \rangle = \frac{m_X}{4\pi \hbar^2 m_X^2} I,
\]

(C3)

where the subscripts “S” and “N” indicate that the change of the energy is taken under adiabatic condition at a given number of particles. Therefore, we obtain the adiabatic relation,

\[
\left( \frac{\partial E}{\partial \ln a_s} \right)_{S,N} = \frac{\hbar^2}{4\pi m_X} I.
\]

(C4)

If we consider the grand-canonical ensemble, where the chemical potential is fixed, by using standard thermodynamic relations, we can re-cast Tan’s adiabatic relation into the form,

\[
\left( \frac{\partial \Omega}{\partial \ln a_s} \right)_{S,\mu} = \frac{\hbar^2}{4\pi m_X} I.
\]

(C5)

By using the thermodynamic potential within the Bogoliubov approximation, i.e., Eq. (A39), we immediately obtain the contact coefficient predicted by the Bogoliubov theory:

\[
I = \frac{m_X^2}{\hbar^4} \left[ \mu + \frac{\Omega^2}{\delta - \mu} \right]^2.
\]

(C6)

Let us now examine the kinetic energy \( \mathcal{F}_X \) and interaction energy \( \epsilon_{\text{int}} \) of excitons,

\[
\mathcal{F}_X + \epsilon_{\text{int}} = \sum_q \frac{\hbar^2 q^2}{2m_X} n_X(q) + \frac{g^2}{2g} \int dr \langle X^\dagger(r)X^\dagger(r)X(r)X(r) \rangle,
\]

(C7)

\[
= \sum_q \frac{\hbar^2 q^2}{2m_X} n_X(q) + \left\{ \frac{m_X}{8\pi \hbar^2} \ln \left[ \frac{4}{e^{2\gamma} m_X a_s^2 \varepsilon_c} \right] - \frac{1}{2} \sum_q \left[ \frac{\hbar^2 q^2}{m_X} + \varepsilon_c \right]^{-1} \right\} \frac{4}{m_X^2} \hbar^4 \mathcal{I}.
\]

(C8)

We may take the infrared cut-off energy \( \varepsilon_c = \hbar^2/ (m_X a_s^2) \) to simplify the equation. This leads to Tan’s energy relation,

\[
\mathcal{F}_X + \epsilon_{\text{int}} = \sum_q \frac{\hbar^2 q^2}{2m_X} \left[ n_X(q) - \frac{I}{q^2 (q^2 + a_s^2)} \right] + \frac{(\ln 2 - \gamma) \hbar^2 I}{4\pi m_X},
\]

(C9)

It is clear from the energy relation that the excitonic momentum distribution must have a universal \( q^{-4} \) tail:

\[
n_X(q \to \infty) = \frac{I}{q^2 (q^2 + a_s^2)} \sim \frac{I}{q^4}.
\]

(C10)
1. The momentum distribution of photons and excitons

One may wonder that the photonic momentum distribution \( n_{\text{ph}}(q) \) may similarly develop a universal \( q^{-4} \) tail, as naively anticipated from the scenario of polariton quasiparticles. However, as we examine directly in the following, this is not the case. The absence of a universal tail in \( n_{\text{ph}}(q) \) is understandable, since it is a large-momentum, high-energy behavior, which can not be captured by the low-energy quasiparticle picture.

To see this, let us calculate the momentum distribution of photons and excitons within the Bogoliubov theory. The Green function is given by,

\[
\mathcal{G}(q, i\nu_n) = \mathcal{G}_{11}(q, i\nu_n) = \frac{(i\nu_n)^3 + A_q (i\nu_n)^3 - (B_q^2 - C^2 - \Omega^2) i\nu_n - A_q (B_q^2 - C^2) - B_q \Omega^2}{(i\nu_n)^2 - E_{q^+}^2} \frac{1}{(i\nu_n)^2 - E_{q^-}^2}.
\]

Integrating over the bosonic Matsubara frequencies \( i\nu_n \), we obtain,

\[
n_{\text{ph}}(q) = \frac{1}{2} \left[ \frac{A_q}{E_{q^+} + E_{q^-}} + \frac{A_q (B_q^2 - C^2) - B_q \Omega^2}{E_{q^+} + E_{q^-}} \right] = \mathcal{O} (q^{-6}).
\]

At large momentum, both \( A_q \) and \( B_q \) are much larger than \( C \) and \( \Omega \). We may use Eq. \( \text{(A27)} \) and Eq. \( \text{(A28)} \) to perturbatively expand \( E_{q^\pm} \). Thus, we find that, when \( q \to \infty \),

\[
n_{\text{ph}}(q) = \frac{C^2}{2 (A_q + B_q)} \frac{A_q (B_q^2 - C^2) - B_q \Omega^2}{2 (A_q + B_q) B_q} = \mathcal{O} (q^{-4}).
\]

Therefore, we conclude that within the Bogoliubov theory, there is no \( q^{-4} \) tail in the photonic momentum distribution. For the excitonic momentum distribution, the Green function of excitons takes the form,

\[
\mathcal{G}_X(q, i\nu_n) = \mathcal{G}_{33}(q, i\nu_n) = \frac{(i\nu_n)^3 + B_q (i\nu_n)^3 - (A_q^2 + \Omega^2) i\nu_n - A_q B_q \Omega^2}{(i\nu_n)^2 - E_{q^+}^2} \frac{1}{(i\nu_n)^2 - E_{q^-}^2},
\]

and the momentum distribution is,

\[
n_X(q) = \frac{1}{2} \left[ \frac{B_q}{E_{q^+} + E_{q^-}} + \frac{A_q B_q - \Omega^2}{E_{q^+} + E_{q^-}} \right] = \mathcal{O} (q^{-4}).
\]

Let us similarly express \( E_{q^\pm} \) in terms of \( A_q \) and \( B_q \) in the large momentum limit. We obtain, for \( q \to \infty \),

\[
n_X(q) = \frac{C^2}{4 (A_q + B_q)^2} \left[ 1 + \frac{A_q B_q}{B_q^2} + \mathcal{O} (q^{-2}) \right] = \frac{C^2}{4 B_q^2} \mathcal{O} (q^{-6}) \approx \frac{m^2}{\hbar^2} \left[ \mu + \frac{\Omega^2}{\delta - \mu} \right]^2 q^{-4}.
\]

Therefore, the contact coefficient extracted from the tail of \( n_X(q) \) is the same as that calculated using the adiabatic relation, see Eq. \( \text{(C6)} \).

Appendix D: Multiple quantum wells

In semiconductor quantum wells, such as GaAs, multiple quantum wells are used to enhance the light-matter coupling [32]. Here, we show that the same results of the polariton-polariton interaction strength and Tan contact coefficient can be derived, up to a trivial factor of \( N \), where \( N \) is the number of quantum wells.
In the presence of $N$ quantum wells, the bosonic model Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{LM}} + \mathcal{H}_{\text{int}}$ takes the form,

$$\mathcal{H}_0 = \sum_{q} \left( \frac{\hbar^2 q^2}{2m_{\text{ph}}} + \delta - \mu \right) \phi_q^\dagger \phi_q + \sum_{i=1}^{N} \sum_{q} \xi_q X_{iq}^\dagger X_{iq}, \quad (D1)$$

$$\mathcal{H}_{\text{LM}} = \frac{\Omega}{\sqrt{NS}} \sum_{i=1}^{N} \sum_{q} \left[ \phi_q^\dagger X_{iq} + X_{iq}^\dagger \phi_q \right], \quad (D2)$$

$$\mathcal{H}_{\text{int}} = \frac{g}{2S} \sum_{i=1}^{N} \sum_{qq'k} X_{i\frac{1}{2}+q}^\dagger X_{i\frac{1}{2}-q}^\dagger X_{i\frac{1}{2}-q'} X_{i\frac{1}{2}+q'}. \quad (D3)$$

Here, $i = 1, ..., N$ is the index of the quantum wells. Each quantum well is assumed to be identical and couples to the cavity with the same light-matter coupling strength $\Omega/\sqrt{N}$.

As before, we assume that photon field and exciton fields condensate at the zero-momentum states with condensate wave-functions $\phi_0$ and $X_0$. At the mean-field level, the thermodynamic potential is

$$\Omega_0 (\mu) = (\delta - \mu) \phi_0^2 + 2\Omega \phi_0 X_0 - \mu X_0^2 + \frac{g}{2N} \tilde{X}_0^4, \quad (D4)$$

which takes the same form as in the case of single quantum well, after we introduce $\tilde{X}_0^2 \equiv N X_0^2$. By minimizing the mean-field thermodynamic potential with respect to $\phi_0$ and $X_0$, we obtain,

$$gX_0^2 = \frac{g}{N} \tilde{X}_0^2 = \mu + \frac{\Omega^2}{\delta - \mu}, \quad (D5)$$

and

$$\Omega_0 = -N \frac{1}{2g} \left[ \mu + \frac{\Omega^2}{\delta - \mu} \right]^2. \quad (D6)$$

Beyond mean-field, we keep the bilinear terms in the field operators and obtain the Bogoliubov action,

$$\mathcal{H}_{\text{Bog}} = \sum_{Q=(q,v_n)} \left[ \delta \phi_Q^\dagger, \delta \phi_{-Q}, \delta X_{1,Q}^\dagger, ..., \delta X_{N,Q}, \delta X_{1,-Q}, ..., \delta X_{N,-Q} \right] \left[ -\mathcal{G}^{-1} (Q) \right], \quad (D7)$$

where the inverse Green function,

$$-\mathcal{G}^{-1} (Q) = \begin{bmatrix} -iv_n + A_q & 0 & \frac{\Omega}{\sqrt{N}} & \cdots & \frac{\Omega}{\sqrt{N}} & 0 & \cdots & 0 \\ 0 & iv_n + A_q & 0 & \cdots & 0 & \frac{\Omega}{\sqrt{N}} & \cdots & \frac{\Omega}{\sqrt{N}} \\ \frac{\Omega}{\sqrt{N}} & 0 & -iv_n + B_q & 0 & 0 & 0 & \cdots & 0 \\ \vdots & \vdots & 0 & \cdots & 0 & 0 & \cdots & 0 \\ \frac{\Omega}{\sqrt{N}} & 0 & 0 & 0 & -iv_n + B_q & 0 & 0 & C \\ 0 & \frac{\Omega}{\sqrt{N}} & C & 0 & 0 & iv_n + B_q & 0 & 0 \\ \vdots & \vdots & 0 & \cdots & 0 & 0 & \cdots & 0 \\ 0 & \frac{\Omega}{\sqrt{N}} & 0 & 0 & C & 0 & 0 & iv_n + B_q \end{bmatrix}. \quad (D8)$$

By solving the poles of the Green function, we find that there is $N - 1$ degenerate eigenvalues

$$E_q = \sqrt{B_q^2 - C^2}, \quad (D9)$$
in addition to the eigenvalues $E_{q+}$ and $E_{q-}$. Therefore, the fluctuation thermodynamic potential is given by,

$$\delta\Omega_g^{(T=0)} = \frac{1}{2} \sum_q \left[ E_{q+} + E_{q-} + (N-1) \sqrt{B_q^2 - C^2 - A_q - N B_q} \right]. \quad (D10)$$

By adding the two thermodynamic potentials and removing the bare interaction strength $g$, we obtain,

$$\Omega = -N m_X C^2 \frac{8\pi \hbar^2}{8\pi \hbar^2} \ln \left[ \frac{4}{e^{2\gamma} m_X a_s^2 \tilde{\varepsilon}_c} \right] + \frac{1}{2} \sum_q \left[ E_{q+} + E_{q-} + (N-1) \sqrt{B_q^2 - C^2 - A_q - N B_q + \frac{N C^2}{\hbar^2 q^2 / m_X + \varepsilon_c}} \right]. \quad (D11)$$

By repeating the steps in Appendix A, in the limit of an infinite mass ratio, it is easy to see that,

$$\sum_q \left[ E_{q+} + E_{q-} + (N-1) \sqrt{B_q^2 - C^2 - A_q - N B_q + \frac{N C^2}{\hbar^2 q^2 / m_X + \varepsilon_c}} \right] = N m_X C^2 \frac{4}{4\pi \hbar^2} \ln \left[ \frac{2 \Omega^2 / (\delta - \mu)}{\varepsilon_c} \right]. \quad (D12)$$

Therefore, we obtain

$$\Omega = -N \frac{m_X C^2}{8\pi \hbar^2} \left[ \frac{\mu + \frac{\Omega^2}{\delta - \mu}}{\frac{\Omega^2}{\delta - \mu}} \right] \ln \left[ \frac{2 \hbar^2 (\delta - \mu)}{e^{2\gamma} m_X a_s^2 \Omega^2 / \tilde{\varepsilon}_c} \right]. \quad (D13)$$

It is readily seen that the thermodynamic potential is trivially enlarged by a factor of $N$, in the case of $N$ quantum wells. As a result, the density is enlarged by $N$ times at a given chemical potential $\mu$ and hence the polariton-polariton interaction strength is reduced by a factor of $N$, i.e.,

$$g_{PP} = X_{LP}^2 \frac{1}{N} \left( \frac{4\pi \hbar^2}{m_X} \right) \ln^{-1} \left[ \frac{2 \hbar^2}{e^{2\gamma} m_X a_s^2 |E_{LP}|} \right]. \quad (D14)$$

In line with this factor of $N$ scaling, Tan contact coefficient within the Bogoliubov theory is now given by,

$$I = \frac{1}{N^2 \ln^2 \left[ e^{2\gamma} m_X a_s^2 |E_{LP}| / (2\hbar^2) \right]} \left( \frac{16\pi^2 n^2}{X_{LP}^4} \right), \quad (D15)$$

which is reduced by a factor of $1/N^2$. 

