

Excitonic oscillator-strength saturation dominates polariton-polariton interactions

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Exciton-polaritons in semiconductor microcavities exhibit large two-body interactions that, thanks to ever-refined nanotechnology techniques, are getting closer and closer to the quantum regime where single-photon nonlinearities start being relevant. To foster additional progress in this direction, in this work we experimentally investigate the microscopic mechanism driving polariton-polariton interactions. We measure the dispersion relation of the collective excitations that are thermally generated on top of a coherent fluid of interacting lower polaritons. By comparing the measurements with the Bogoliubov theory over both the lower- and upper-polariton branches simultaneously, we find that polariton-polariton interactions stem dominantly from a mechanism of saturation of the exciton oscillator strength.

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In the broad context of fundamental and applied quantum sciences, visible photons constitute versatile and powerful quantum degrees of freedom [1–3]. Quantum states of light can indeed be prepared and sent over long distances with minimal losses in free space [4–6] or in optical-fiber networks [7–9] for quantum communication, and manipulated in advanced photonic circuits to perform quantum computing tasks [10–12]. These capabilities are enabled by the fact that visible photons can be detected individually with a high quantum efficiency, and by the fact that they can be spatially and spectrally manipulated by engineering the dielectric environment [13–15]. But unlike other quantum systems such as superconducting circuits, trapped ions, or ultracold atoms, making visible photons to strongly interact with each other [16] remains a daunting challenge for practical and scalable solid-state systems.

In recent years, significant progress in this direction has been made by engineering effective interactions between photons propagating in solid-state systems, in order to dress photons with a suitable electronic transition from, e.g., a

quantum dot [17–20] or a quantum well [21–24]. In the latter case, when the quantum well is embedded in a microcavity, the system can operate in the strong coupling regime between excitons and photons leading to exciton-polaritons (in short, polaritons). The dressing thus exceeds the perturbative regime and the resulting exciton-mediated two-body interaction is dramatically enhanced.

In GaAs-based open-microcavity systems [25], a polariton-polariton interaction figure of merit $\mathcal{F} = U/\gamma \simeq 0.15$ has been reported [21,22], where $\hbar U$ is the interaction strength between two polaritons and $\hbar\gamma$ is the polariton spectral linewidth. In order to enter the quantum regime $\mathcal{F} \gtrsim 1$, different strategies are currently studied, such as adding a static dipole moment to excitons to increase Coulomb interactions [26–29], coupling polaritons to a sea of free charges to obtain polaron-polaritons [30] or quantum Hall polaritons [31], using large Rydberg excitons with high principal quantum number [32], or exploiting biexcitonic Feshbach resonances in the two-polariton scattering [23,33–35]. A major difficulty along this path is that a precise understanding of the mechanism dominating polariton-polariton interactions is still lacking, even in the simplest configurations.

Early experiments in bare quantum wells suggest that two-body scattering plays an important role in the interactions between bare excitons [36]. However, due to a very different regime of coherence and of light-matter coupling strength [37–40], qualitative and quantitative differences are expected for the case of polariton-polariton interactions. In fact, a growing number of experiments in

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GaAs-based [41–43] and in transition metal dichalcogenide (TMD)-monolayer microcavities [28,44,45] indicate that a saturation mechanism of the excitonic oscillator strength might significantly contribute to the polariton-polariton interaction.

In order to understand this unexpected feature in full generality, here we develop a dedicated method to accurately discriminate between saturation mechanism and exciton-exciton scattering. We investigate a typical GaAs-based microcavity over a large range of exciton-photon detuning and of interaction energy, and we measure the dispersion relation of the collective Bogoliubov excitations of the coherent polariton fluid [46]. When upper- and lower-polariton excitations are simultaneously considered, we demonstrate that they provide quantitative information on the mechanism governing polariton-polariton interactions. For all investigated parameters, we find that polariton-polariton interaction is dominated by a saturation mechanism of the excitonic oscillator strength, and we rule out other plausible explanations.

In order to model interacting polaritons in our experiment, we consider strongly coupled excitons and photons confined in a planar microcavity, described by the Hamiltonian

$$\hat{\mathcal{H}} = \hbar \sum_{\mathbf{k}} \left[\omega_x \hat{b}_{\mathbf{k}}^\dagger \hat{b}_{\mathbf{k}} + \omega_{c,\mathbf{k}} \hat{a}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}} + \frac{\Omega}{2} (\hat{a}_{\mathbf{k}}^\dagger \hat{b}_{\mathbf{k}} + \text{H.c.}) \right] + \hat{F} + \hat{\mathcal{H}}_{xx} + \hat{\mathcal{H}}_{xc}, \quad (1)$$

where $\hat{b}_{\mathbf{k}}^\dagger$ and $\hat{a}_{\mathbf{k}}^\dagger$ are the bosonic creation operators for excitons and photons, respectively, with in-plane momentum $\hbar\mathbf{k}$, coupled to each other at a Rabi frequency $\Omega/2$, and driven resonantly by a coherent field of in-plane momentum $\hbar\mathbf{k}_p$ coupled to cavity photons as $\hat{F} = (\hat{a}_{\mathbf{k}_p} F_0^* e^{i\omega_{\text{las}} t} + \text{H.c.})$. The excitonic transition frequency ω_x can be safely considered constant inside the polariton-relevant light cone, and the cavity-mode dispersion relation has the form $\omega_{c,\mathbf{k}}^2 = \omega_{c,0}^2 + c^2 k^2 / n_{\text{cav}}^2$, where n_{cav} is the effective refractive index of the cavity mode.

The interactions terms $\hat{\mathcal{H}}_{xx}$ and $\hat{\mathcal{H}}_{xc}$ constitute the main focus of this work. The exciton-exciton scattering $\hat{\mathcal{H}}_{xx}$ is modeled by

$$\hat{\mathcal{H}}_{xx} = \frac{\hbar g_{xx}}{2} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} \hat{b}_{\mathbf{k}+\mathbf{q}}^\dagger \hat{b}_{\mathbf{k}'-\mathbf{q}}^\dagger \hat{b}_{\mathbf{k}} \hat{b}_{\mathbf{k}'} \quad (2)$$

with coupling strength $\hbar g_{xx}$ [47]. In Ref. [48], $\hbar g_{xx}$ was determined to be approximately constant in momentum space within the light cone, and with a magnitude determined by the carrier-exchange contribution to Coulomb interaction. When the exciton-photon coupling is included in the model, either in the weak [49] or in the strong coupling regime relevant for polaritons [50,51], another interaction mechanism is found, the excitonic saturation $\hat{\mathcal{H}}_{xc}$, which is rooted into the fermionic nature of the electrons and holes constituting excitons, and reads [49–51]

$$\hat{\mathcal{H}}_{xc} = -\frac{\hbar g_{xc}}{2} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} (\hat{a}_{\mathbf{k}+\mathbf{q}}^\dagger \hat{b}_{\mathbf{k}'-\mathbf{q}}^\dagger \hat{b}_{\mathbf{k}} \hat{b}_{\mathbf{k}'} + \text{H.c.}), \quad (3)$$

where $\hbar g_{xc}$ is the interaction strength. The main effect of this term is to reduce the effective exciton-photon coupling [of

nominal strength $\hbar\Omega$ in Eq. (1)] as the excitonic density n_x increases.

Owing to the relatively low polariton density involved in the experiment, we can work within a mean-field approximation. Following Ref. [43], the dispersion relation of the Bogoliubov excitations can thus be obtained by expanding to quadratic order the Hamiltonian (1) on top of a polaritonic mean field and then diagonalizing the Bogoliubov matrix $\hat{M}_B(\mathbf{k}_p, \mathbf{k})$ (see Sec. II of the Supplemental Material [52]). The excitation spectrum displays four branches $\omega_{B,j}(k)$: the lower-polariton ghost (labeled lg) and normal (labeled ln) branches, and the upper-polariton ghost (labeled ug) and normal branches (labeled un).

A key characteristic of these branches is the fact that their shape and relative spectral shift depend significantly on the relative contribution of the exciton-exciton (2) and saturation (3) interaction mechanisms [52]. When H_{xx} dominates, the effective excitonic transition of the Bogoliubov states blueshift and hence both ln and un blueshift by a comparable amount. When H_{xc} dominates, the effective excitonic transition also blueshifts (see Sec. II B 1 of the Supplemental Material [52]), but, furthermore, the effective Rabi splitting is reduced. For ln these two corrections add up into a blueshift, while they contribute with an opposite sign for un, which is thus subjected to a much weaker spectral shift, which changes sign around $\delta_{xc} = 0$. This difference is a key observable to pinpoint the relative contributions of the H_{xx} and H_{xc} interaction mechanisms from the measurement of both $\omega_{B,\text{ln}}(k)$ and $\omega_{B,\text{un}}(k)$.

In the experiment, we employ a state-of-the-art planar GaAs-based microcavity (identical to that in Ref. [43]) operating in the strong coupling regime between the cavity photons and the excitonic transition of a single high-quality 17 nm thick In_{5%}Ga_{95%}As quantum well embedded in the cavity spacer. This spacer is intentionally wedge-shaped, so that the photon-exciton detuning $\delta_{xc} = \hbar(\omega_c(k=0) - \omega_x)$ can be chosen at will by changing the working point at the cavity surface. The sample is operated at $T = 3.9$ K. Polaritons are excited using a continuous wave laser focused into a Gaussian spot of 35 μm radius, of which only the central part of radius $R_{\text{det}} = 15 \mu\text{m} \gg \xi$ is kept for detection. For the chosen experimental parameters, $\xi \simeq \hbar/\sqrt{2mE_{nl}} < 3.5 \mu\text{m}$ is the healing length of the lower-polariton quantum fluid, and the characteristic energy scale of the interactions in the polaritonic fluid is varied in the range $E_{nl} = \hbar(g_{xx} + g_{xc})n_x \in [50, 300] \mu\text{eV}$ by varying the driving laser intensity. A weak polaritonic disorder is present within the excitation spot but its amplitude, much smaller than the linewidth, is found to leave the dispersion relation unperturbed. The laser frequency is tuned either (1) far above the polaritonic resonances to excite low-density polariton photoluminescence [Figs. 1(a) and 1(c)] or (2) close to resonance with the lowest polariton state at $k_p \simeq 0$ [Figs. 1(b) and 1(d)]. We have verified that in condition (2), residual heating from the laser does not induce any measurable excitonic thermal redshift [52]. The laser polarization is chosen circular in order to focus on the interaction between copolarized polaritons. The interaction between cross-polarized polaritons is significantly smaller and involves different physical mechanisms such as biexciton Feshbach enhancement [23,34,35,53] and intermediate spin-2 exciton states [54]: its study goes

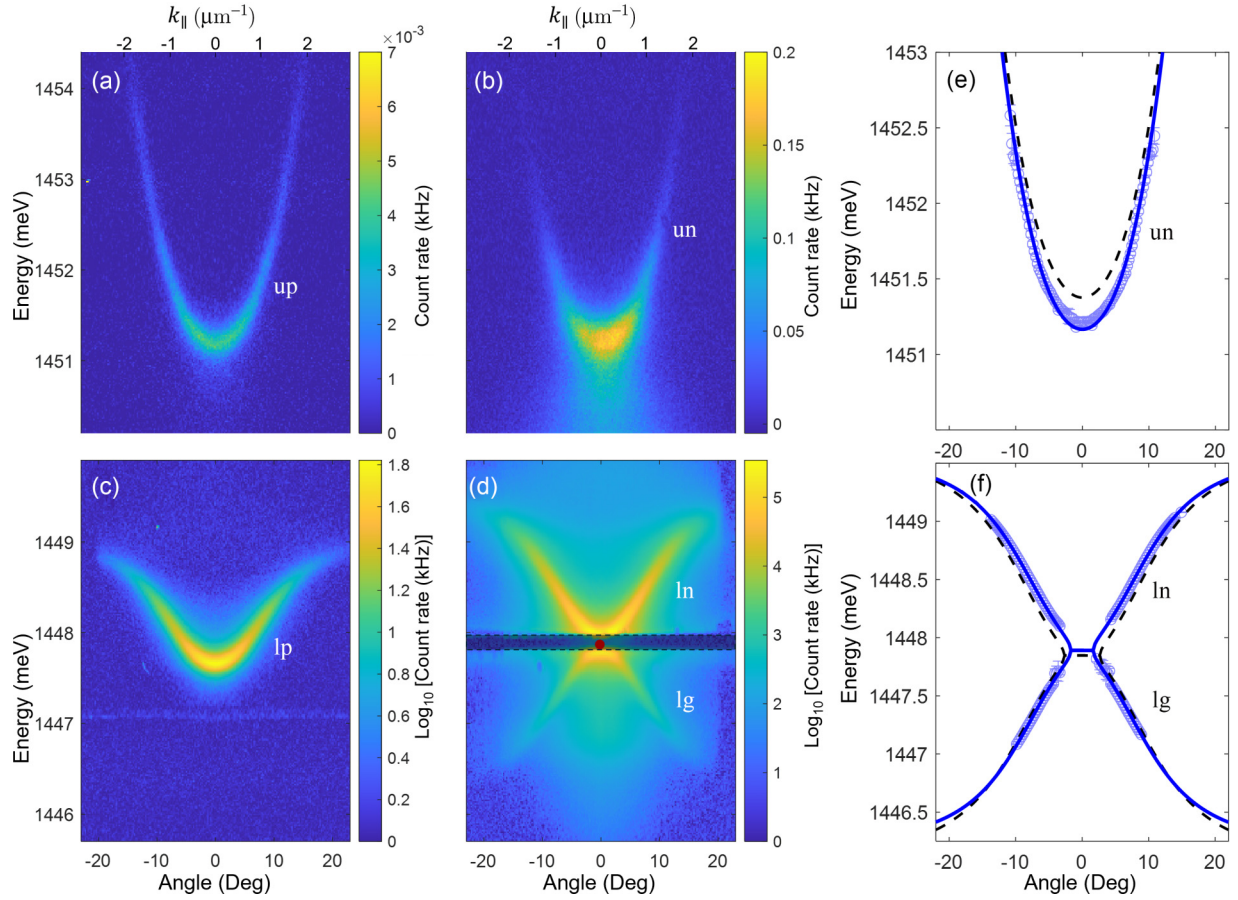


FIG. 1. Analysis of working point WP3c (see main text). Left panels: Photoluminescence maps $I_{PL}(\theta, \omega)$ of (a) the upper- and (c) the lower-polariton branches obtained under weak nonresonant excitation (with vanishing two-body interactions). Central panels: Emission map $I_B(\theta, \omega)$ of (b) the upper-polariton normal (un), and (d) lower-polariton normal (ln) and ghost (lg) (negative curvature) Bogoliubov excitation branches under resonant excitation. The dashed rectangle in panel (d) shows where the mean-field emission is rejected by the spectral filter, and the red circle indicates ω_{las} . Right panels: Measured and fitted dispersion relation of (e) the un and (f) the ln and lg branches. The blue symbols are the measurements obtained from analysis of the data in panels (b) and (d). The solid blue (dashed black) line is a fit using Bogoliubov theory, and leaving $g_{xx}n_x$ and $g_{xc}n_x$ as free parameters (leaving $g_{xx}n_x$ as free parameter, and setting $g_{xc}n_x = 0$). See main text for details.

beyond the scope of this Letter and will be examined in a future work.

For each investigated working point (WP), we first use the laser setting (1) to collect angle- and energy-resolved polariton photoluminescence maps $I_{PL}(\theta, \omega)$ from which the physical photoluminescence maps of each investigated working point can be accurately determined (see Sec. I of the Supplemental Material [52]). The measurement is shown in Figs. 1(a) and 1(c) for WP3. We then use the resonant laser setting (2) where we apply different, slightly blue-detuned laser frequencies ω_{las} with respect to the noninteracting lower-polariton state frequency $\omega_{0,lp}(0)$: $\delta_{las} = \omega_{las} - \omega_{0,lp}(k=0) \in [50, 300] \mu\text{eV}$. For all investigated WPs, the blue-detuned δ_{las} is chosen to be large enough for the polaritonic field to be bistable [46]. We then drive the system using different values of laser power and hence of polaritonic densities, in the high-density branch of the bistability, that offers optimal conditions to generate a stable steady-state polaritonic fluid with an interaction energy comparable to or larger than $\hbar\delta_{las}$ [43,55]. The transmitted coherent light is then rejected by narrow-band spectral filtering, in order to collect only the emission $I_B(\theta, \omega)$ from the Bogoliubov excitations that are dominantly generated by coupling to

the thermal phonons of the solid-state environment [43]. The Bogoliubov emission $I_B(\theta, \omega)$ collected in this way at WP3, for $\delta_{las} = 203 \mu\text{eV}$ and a laser power of 13.7 mW, is shown in Figs. 1(b) and 1(d) (experiment labeled “WP3c” thereafter). A list of all investigated experiments “WP_{jk}” can be found in Table II of the Supplemental Material [52]. For all WPs, these parameters are chosen for the system to be close to the sonic regime of the collective excitations [46]. For each angle θ , $I_B(\theta, \omega)$ exhibits three resonance peaks corresponding—in increasing energy order—to the lg, ln, and un Bogoliubov states; note that the ug resonance yields a too weak emission to be observed in these experimental conditions. The measured dispersion relations extracted from $I_B(\theta, \omega)$ are shown in Figs. 1(e) and 1(f) for WP3c (blue symbols).

The comparison of these measured dispersion relations with the predictions of the Bogoliubov theory is realized as follows. As we have already determined the linear regime parameters for each WP and we know the value of the applied laser detuning δ_{las} , the only free parameters of the model are the two nonlinear energies $E_{nl,xx} = g_{xx}n_x$ and $E_{nl,xc} = g_{xc}n_x$ that the fitting algorithm is tasked to determine. It consists in a least-squares optimization algorithm that is simultaneously

applied to all points along the three measured dispersion relation branches. In doing so, our main target is to determine the relative contribution of g_{xc} to the polariton-polariton interaction, defined as $R_{xc} = g_{xc}/(g_{xx} + g_{xc}) = E_{nl,xc}/(E_{nl,xx} + E_{nl,xc})$. Importantly, we do not need to determine the absolute value of g_{xx} or g_{xc} independently, which would require the difficult task of determining the excitonic density n_x in absolute terms.

The theoretical dispersion relations obtained in this way are shown in Figs. 1(e) and 1(f) for WP3c as the solid blue line. We quantify the goodness of fit using the root-mean-squared error S_{fit} where $S_{\text{fit}}^2 = \langle (\omega_{j,\text{exp}} - \omega_{j,\text{fit}})^2 \rangle_j$, where $\langle \cdot \rangle_j$ is an average running over the experimental points θ_j . For WP3c, $S_{\text{fit}} = 27.4 \mu\text{eV}$, and the obtained parameters are $E_{nl,xc} = 188 \pm 12 \mu\text{eV}$ and $E_{nl,xx} = 13 \pm 17 \mu\text{eV}$, which leads to $R_{xc} = 94\% \pm 8\%$, where the uncertainty is the 1σ confidence interval. Fitting with H_{xc} only, and hence with a single free parameter $g_{xc}n_x$, yields an excellent agreement as well, of $S_{\text{fit}} = 28 \mu\text{eV}$ (see Table VI of the Supplemental Material [52]). The saturation mechanism is thus strongly dominant over the exciton-exciton interaction. For comparison, we also fitted WP3c assuming $g_{xc} = 0$, i.e., keeping only the exciton-exciton $\hat{H}_{xx}$ interaction term. The result is shown as the black dashed line in Figs. 1(e) and 1(f) and yields an agreement of $S_{\text{fit}} = 140 \mu\text{eV}$, i.e., $5\times$ poorer. We performed this analysis for all investigated WPs in which $\delta_{xc} = \{-0.25; 0.10; 0.30; 0.53; 1.55\}$ meV and different parameter sets in terms of laser detuning δ_{las} and the characteristic interaction energy $E_{nl} \in [50, 300] \mu\text{eV}$ (see Table III of the Supplemental Material for the results [52]). The resulting values of R_{xc} are summarized in Fig. 2(a): These results systematically show a large domination of the saturation mechanism [Eq. (3)] over the exciton-exciton interaction [Eq. (2)]. Its average contribution thus amounts to $\langle R_{xc} \rangle = 96\% \pm 11\%$.

We confirm this conclusion using a second method that does not require any fitting operation, and which is focused on the particularly high sensitivity of the upper-polariton Bogoliubov branch energy shift to the different interaction contributions. We first determine $\Delta\omega_l = \omega_{B,\text{ln}}(\theta_l) - \omega_{0,\text{lp}}(\theta_l)$ as the ln branch spectral shift, where $\theta_l = 9^\circ$ is chosen such that $\omega_{B,\text{ln}}(\theta_l) > E_{nl}/\hbar$ and hence the measured spectral shift is not influenced by the particle-hole mixing. On the un branch, this condition is verified at all angles, so we define $\Delta\omega_u = \omega_{B,\text{un}}(0) - \omega_{0,\text{up}}(0)$, as $\theta_u = 0$ yields the best signal-to-noise ratio. The ratio $\rho_B = \Delta\omega_u/\Delta\omega_l$ is plotted in Fig. 2(b) (blue symbols) as a function of the exciton-photon detuning δ_{xc} and compared to the Bogoliubov theory assuming $R_{xc} = \langle R_{xc} \rangle$ for all points (solid blue line) and $R_{xc} = 0$ (solid green line). While the exciton-exciton scattering model is distant from the experimental data points by more than $4\times$ the experimental 1σ confidence interval, the agreement with a saturation-mechanism-dominated interaction is again striking: In particular, the sign change of ρ_B around $\delta_{xc} \simeq 0$ is a strong indicator of the compensation mechanism between $\tilde{\Omega}$ and $\tilde{\omega}_x$ that occurs for the upper-polariton branch when a saturation mechanism dominates as discussed earlier. Within our experimental resolution, we checked that the dependence of ρ_B on E_{nl} is also qualitatively consistent with our mean-field model dominated by a saturation mechanism [cf. Fig. 2(c)].

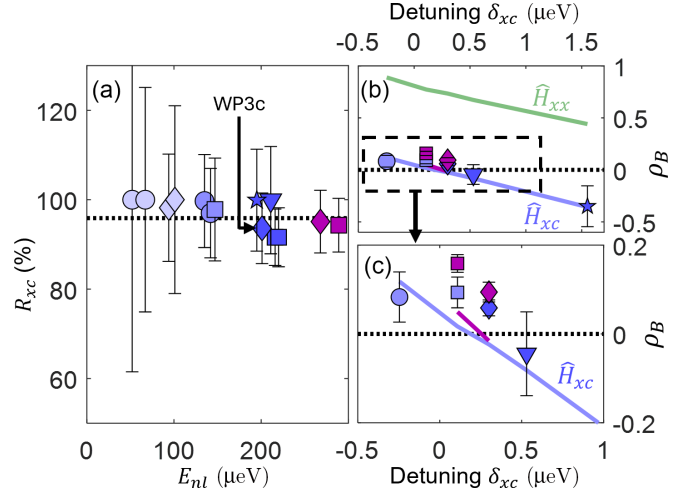


FIG. 2. (a) Contribution of the saturation mechanism $R_{xc} = g_{xc}/(g_{xc} + g_{xx})$ to the polariton-polariton interaction as a function of the characteristic interaction energy E_{nl} . Five WPs with exciton-photon detunings $\delta_{xc} = -0.25, +0.10, +0.30, +0.53$, and $+1.55$ meV are plotted as circles, squares, diamonds, triangle, and star symbols, respectively. For each WP, the lowest to highest probed E_{nl} are plotted from light to dark shades of blue (purple for the highest). The dashed black line shows the average ratio $\langle R_{xc} \rangle = 96\%$. The arrow indicates WP3c. (b) Upper- to lower-polariton spectral shift ratio ρ_B as a function of δ_{xc} using the same symbols and color notations. A single parameter set per WP is plotted for clarity: WP1c, WP2a, WP2d, WP3c, WP3d, WP4a, and WP5a (see Table II of the Supplemental Material [52]). The thick blue (green) line is the Bogoliubov theory with $R_{xc} = \langle R_{xc} \rangle$ ($R_{xc} = 0\%$). Panel (c) is a closeup of panel (b). WP2d and WP3d (purple color) are different from WP2a and WP3c (blue) only by their higher E_{nl} . The theory for WP2d and WP3d is the solid purple line segment.

Finally, we checked how our observations compare with other plausible interaction models, namely, an additional excitonic thermal redshift, the scattering of polaritons with a residual incoherent reservoir of dark excitons [56–58] or laser heating activated free charges [59], an oscillator strength saturation mechanism mediated by such a reservoir [60,61], an exciton-photon scattering mechanism, or a nonlinear contribution to the Rabi coupling. The details of this analysis are given in Sec. III of the Supplemental Material [52] and show that none of these models offers a better fit to the data than H_{xc} . This analysis also shows that both H_{xx} and the interaction with an incoherent reservoir can possibly account for the remaining 5% contribution to the polariton-polariton interaction.

We highlight that our conclusion is fully compatible with previous works where only the exciton-exciton interaction term $\hat{H}_{xx}$ was used to model polariton-polariton interactions: These works focused on the lower-polariton dispersion branch only, in which both terms $\hat{H}_{xx}$ and $\hat{H}_{xc}$ lead to repulsive lp-lp interactions, which is the key feature in polaritonic quantum fluid phenomena. Only this dedicated analysis—that crucially involves the upper-polariton branch—has allowed us to clearly distinguish between these two mechanisms.

Note also that in this work we have not made any assumptions on the microscopic mechanisms underlying the H_{xx} and H_{xc} Hamiltonian terms. To shed light on them, and understand, for instance, whether our conclusions applies to other materials or beyond the investigated parameter ranges, different microscopic theories are available in the literature, e.g., accounting for the electron-hole degrees of freedom [62,63] and their coupling to photons [60,64–66]. Interestingly, in Refs. [65,67], the authors observe in their model some features in the lower-polariton sector that are consistent with a saturation mechanism; however, as discussed earlier, a meaningful comparison between experiment and theory needs addressing both the lower- and upper-polariton sectors. In a forthcoming work, we will extend the model developed in Ref. [67] to explicitly include the upper-polariton branch as well as saturation effects, and compare the predictions to the experimental observations.

In conclusion, our results demonstrate that, in a widely used InGaAs-based microcavity design, the polariton-polariton interaction is dominated by excitonic oscillator-strength saturation effects, while exciton-exciton scattering or the effect of an incoherent reservoir contributes to less than 5% of the total interaction energy over a widely used range of exciton-photon detuning and polariton density, which corresponds to an excitonic fraction range $|X|^2 \in [0.46, 0.70]$ and

an interaction energy range $|\hbar(g_{xx} + g_{cx}) \in [50, 300] \mu\text{eV}$. This result establishes a saturation-based picture of polariton-polariton interactions with far-reaching consequences in the quest for reaching the full quantum regime.

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Data availability. The data that support the findings of this article are openly available [68].

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