

Many-Body and Correlation Effects on Parametric Polariton Amplification in Semiconductor Microcavities

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The complexity induced by the Coulomb interaction between electrons determines the noninstantaneous character of exciton-exciton collisions. We show that the exciton-photon coupling in semiconductor microcavities is able to alter the exciton dynamics during collisions strongly affecting the effective scattering rates. Our analysis clarifies the origin of the great enhancement of parametric gain observed when increasing the polariton splitting. It also demonstrates that exciton-exciton collisions in semiconductors can be controlled and engineered to produce almost decoherence-free collisions for the realization of all-optical microscopic devices.

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The phase-coherent amplification of light-matter waves (cavity polaritons) recently observed is one of the most exciting developments in the field of semiconductor nonlinear optics [1–7]. It operates with analogous principles (but under very different conditions) of matter-wave amplifiers based on ultracold atoms [8,9]. Very recently it has been shown that a semiconductor microcavity (SMC) can amplify (via phase-coherent amplification of polaritons) a weak light pulse more than 5000 times [7].

Cavity polaritons are two-dimensional eigenstates of SMCs which result from the strong resonant coupling between cavity-photon modes and two-dimensional excitons in embedded quantum wells (QWs) [10]. The dynamics and hence the resulting energy bands of these mixed quasiparticles are highly distorted with respect to those of bare excitons and cavity photons (Fig. 1). The exciton-photon coupling rate V determines the splitting ($2V$) between the two polariton energy bands. Coherent amplification of polaritons requires a coupling mechanism able to transfer polaritons from a reservoir (in this case provided by polaritons resonantly excited by a pump laser pulse on the lower polariton dispersion) to the signal mode, while conserving energy and momentum. Polaritons of different modes are coupled via their excitonic content, the coupling being provided mainly by the Coulomb interaction between excitons (also the anharmonic part of the exciton-photon interaction contributes). The microscopic theory of parametric polariton amplification [11] shows that the resulting Coulomb coupling strength is given by the exciton-exciton (XX) scattering rate [12] $V_{XX} \approx 1.52E_b a_0^2$ (E_b is the exciton binding energy and a_0 is the exciton Bohr radius) times the exciton fraction of the interacting polariton modes. This effective polariton-polariton interaction scatters a pair of pump polaritons into the lowest-energy state and into a higher-energy state (usually known as the idler mode). Energy and momentum conservation requires $2\omega_k =$

$\omega_0 + \omega_{2k}$, where ω_k is the energy of a pump polariton injected with an in-plane wave vector \mathbf{k} as shown in Fig. 1. The scattering process is stimulated by a weak signal beam injected perpendicular to the cavity ($\mathbf{k} = 0$) that is thus greatly amplified. It has recently been shown that increasing the exciton-photon coupling V by inserting a large number of QWs into the cavity ($V = V_1 \sqrt{N_{\text{eff}}}$, where V_1 is the exciton-photon coupling for

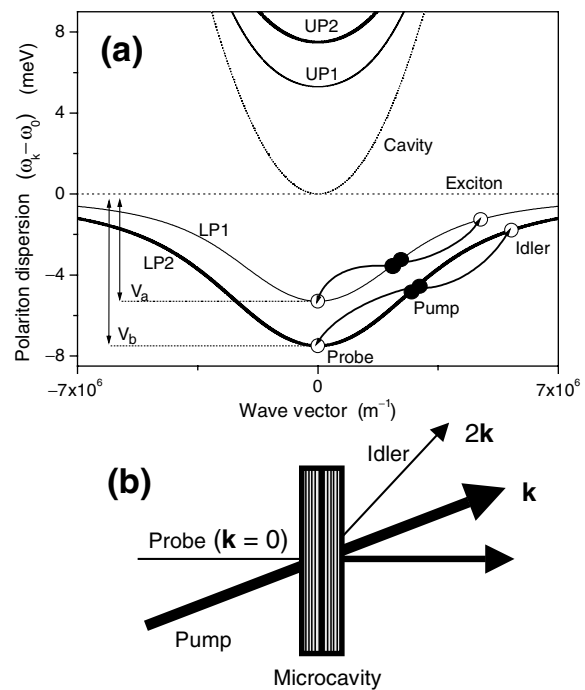


FIG. 1. Sketch of the polariton energy versus the in-plane wave vector (a), for two different exciton-photon coupling ($2V_a = 10.4$ and $2V_b = 15$ meV). UP and LP denote the split upper polariton and lower polariton curves. The parametric polariton-polariton scattering process is also depicted. (b) Schematic of the experiment.

1 QW and the effective number of QWs N_{eff} depends on the number of wells inside the cavity and their spatial overlap with the cavity mode), greatly increases amplification [7]. This spectacular enhancement of gain contrasts with the results of present theories [11,13] describing the process. According to mean-field descriptions, the gain is limited only by the pump depletion and can thus reach very high values (orders of magnitude higher than experimental findings, as shown in this Letter) also for small values of V . Moreover, according to these descriptions, inserting more QWs just lowers the polariton-polariton interaction, without changing the structure of pertinent equations. In particular we observe that the XX interaction in the presence of more QWs results to be V_{XX}/N_{eff} , while the exciton and photon contents of the three interacting modes result in being independent of V . From this observation and from a direct inspection of the pertinent equations, it descends that, increasing the exciton-photon coupling, should not increase the amplification of a weak signal beam in contrast with the spectacular experimental observations [7].

Here we show that the great enhancement of gain, observed when increasing the polariton splitting, has its origins from the unique interplay of the noninstantaneous nature of XX interactions [14] with the strong coupling regime giving rise to polaritons. The mean-field theory of the optical response of electron-hole pairs assumes that collisions are instantaneous and only the mean free time between collisions matters. Although the mean-field theory has enjoyed considerable success [11,14], recent studies revealed that collisions between excitons are more complex (see [14] and references cited therein). They involve multiparticle (at least four-particle) entangled states. This complexity, induced by the Coulomb interaction between electrons, determines the finite duration of collisions [12,14–18]. We find that the strong coupling of excitons with cavity photons, giving rise to polaritons, alters the excitonic dynamics during XX collisions, producing a modification of the effective scattering rates. As a consequence, a variation of the exciton-photon coupling rate V can affect significantly XX collisions and hence the amplification process. To analyze the influence of the finite duration of XX interactions on the parametric polariton amplification, we use a microscopic description where the hierarchy of higher order density matrices has been closed by invoking the dynamics controlled truncation (DCT) scheme [19]. The calculations have been performed in the so-called coherent limit. Furthermore, we included multiple scattering simply by replacing in the nonlinear sources, linear light, and polarization fields with the total fields. The intracavity light field is described within the quasimode scheme [16]. The time evolution of the coupled exciton ($P_{\mathbf{k}}$) and photon waves ($E_{\mathbf{k}}$) including finite duration of XX collisions [16,17] can be described by the following set of coupled equations,

$$\begin{aligned}\frac{\partial}{\partial t} E_{\mathbf{k}} &= -(\gamma_c + i\omega_k^c) E_{\mathbf{k}} + iVP_{\mathbf{k}} + t_c E_{\mathbf{k}}^{\text{in}}, \\ \frac{\partial}{\partial t} P_{\mathbf{k}} &= -(\gamma_x + i\omega_x) P_{\mathbf{k}} + iVE_{\mathbf{k}} - i\Omega_{\mathbf{k}}^{\text{NL}},\end{aligned}\quad (1)$$

where ω_k^c , ω_x , and γ_c , γ_x are the energies and dephasing rates of cavity photons and QWs excitons, respectively. $E_{\mathbf{k}}^{\text{in}}$ describes input light pulses, t_c determines the beam fraction passing the cavity mirror ($t_c = \sqrt{\gamma_c}$ for a cavity with equal mirrors). The intracavity and the exciton field of a given mode \mathbf{k} are coupled by V . The relevant nonlinear source term, able to couple waves with different in-plane wave vector \mathbf{k} [20], is given by $\Omega_{\mathbf{k}}^{\text{NL}} = (\Omega_{\mathbf{k}}^{\text{sat}} + \Omega_{\mathbf{k}}^{\text{XX}})/N_{\text{eff}}$, where the first term originates from the phase-space filling of the exciton transition,

$$\Omega_{\mathbf{k}}^{\text{sat}} = \frac{V}{n_{\text{sat}}} \sum_{\mathbf{k}' \mathbf{k}''} P_{\mathbf{q}}^* P_{\mathbf{k}''} E_{\mathbf{k}'}, \quad (2)$$

being $n_{\text{sat}} = 7/(4\pi a_0^2)$ the exciton saturation density (\mathbf{k}' , \mathbf{k}'' , and \mathbf{q} are tied by the momentum conservation relation $\mathbf{k} + \mathbf{q} = \mathbf{k}' + \mathbf{k}''$). $\Omega_{\mathbf{k}}^{\text{XX}}$ is the Coulomb interaction term. It dominates the coherent XX coupling and for cocircularly polarized waves can be written as

$$\begin{aligned}\Omega_{\mathbf{k}}^{\text{XX}} &= \sum_{\mathbf{k}' \mathbf{k}''} \left[V_{XX} P_{\mathbf{q}}^*(t) P_{\mathbf{k}''}(t) P_{\mathbf{k}'}(t) \right. \\ &\quad \left. - iP_{\mathbf{q}}^*(t) \int_{-\infty}^t F(t-t') P_{\mathbf{k}''}(t') P_{\mathbf{k}'}(t') \right].\end{aligned}\quad (3)$$

$\Omega_{\mathbf{k}}^{\text{XX}}$ includes the instantaneous mean-field XX interaction term (V_{XX}) plus a noninstantaneous term originating from four-particle correlations. This coherent memory can be interpreted as a non-Markovian process involving the 2-particle (excitons) polarization waves interacting with a bath of four-particle correlations [14,15]. The analytical expression of $F(\tau)$ in terms of the two-exciton wave functions can be found in Refs. [12,16,18,19]. We observe that the strong exciton-photon coupling does not modify the memory kernel $F(\tau)$ as a consequence of the fact that four-particle correlations do not couple to cavity photons [16–18]. However, cavity effects are able to alter the phase dynamics of two-particle polarization waves $P_{\mathbf{k}}$ during collisions, i.e., on a time scale shorter than the decay time of the memory kernel $F(\tau)$. In particular, the phase of 2-particle polarization waves $P_{\mathbf{k}}$ in SMCs oscillates with a frequency ω_k (fixed by the polariton dispersion relations) modified respect to that of excitons in bare QWs. This fact produces a modification of the integral in Eq. (3). In this way the exciton-photon coupling V affects the XX collisions that govern the polariton amplification process. This mechanism can be more clearly understood simplifying the integral in Eq. (3). Let us consider a situation where the pump, the signal, and idler energies are all close to the corresponding polariton resonance values and the broadening is small compared to the polariton splitting $2V$. Then it is a good approximation to replace the integral in Eq. (3) with a simpler expression [21], adopting the Weisskopf-Wigner approximation used

to analyze the spontaneous emission associated with atomic transitions. Within this approximation, the dominant XX interaction term for the signal (0) and idler ($2\mathbf{k}$) modes can be written as [17]

$$\Omega_{0(2\mathbf{k})}^{XX} = \mathcal{F}(\omega_{0(2\mathbf{k})} + \omega_{\mathbf{k}})|P_{\mathbf{k}}(t)|^2 P_{0(2\mathbf{k})}(t) + \mathcal{F}(2\omega_{\mathbf{k}})P_{2\mathbf{k}(0)}^*(t)P_{\mathbf{k}}^2(t), \quad (4)$$

where the complex quantity $\mathcal{F}(\omega)$ is the Fourier transform of the memory kernel $F(\tau)$ plus the mean-field contribution V_{XX} . The first term produces a blueshift of the polariton resonance and introduces an intensity-dependent dephasing mechanism. The second term provides the coupling mechanism able to transfer polaritons from the pump to the signal and idler modes. An analogous expression can be derived for the XX interaction of the pump mode. This Weisskopf-Wigner description of four-particle correlation effects in SMCs sets out the relevance of polariton pairs in the scattering process. The energy of these pairs determines the effective scattering rates as shown by Eq. (4). The spectral function $\mathcal{F}(\omega)$ can be calculated by numerical diagonalization of the semiconductor Hamiltonian in the four-particle subspace. We calculated it for QW excitons following a recent microscopic approach [12,18] based on the T matrix (Fig. 2). The obtained four-particle spectral density displays strong variations within the spectral region of interest around $2\omega_0$. In particular, moving towards the low energy region, the dispersive part $\text{Re}(F)$ increases while the absorptive part $\text{Im}(F)$ (it reflects the density of states of 2-exciton correlations [15]) that contrasts gain goes to zero. We observe that the energies of the pump, signal, and idler polaritons lowers with the increase of polariton splitting ($2V$) [see Fig. 1(a)]. The increase of V thus modi-

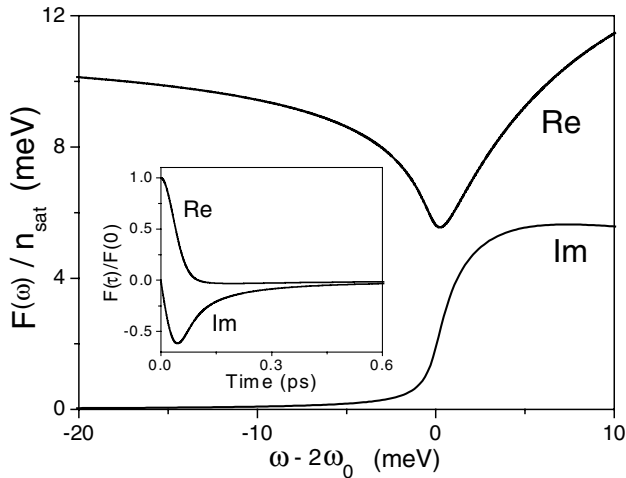


FIG. 2. Energy dependence of the effective exciton-exciton scattering potential. It has been calculated for a GaAs QW 7 nm wide with exciton binding energy 13.5 meV and using as dephasing rate of the four-particle states $\Gamma = 2\gamma_x = 0.58$ meV. The tail of $\text{Im}[\mathcal{F}(\omega)]$ at negative detuning ($\omega < 2\omega_0$) is produced by Γ and vanishes for $\Gamma \rightarrow 0$. The inset shows the dynamics of the memory kernel $F(\tau)$.

fies the effective collision rates in such a way that favors the amplification process. This analysis explains the large increase of gain observed when maximizing the exciton-photon coupling [7]. The gain curve versus the input pump power, obtained solving numerically the system of integro-differential equations (1) for GaAlAs-based samples with splitting $2V_a = 10.6$ meV ($N_{\text{eff}} = 4$) and $2V_b = 15$ meV ($N_{\text{eff}} = 8$) [Fig. 3(a)], fully confirms this analysis. The calculated gain is defined as the total light intensity transmitted in the signal direction divided by the intensity transmitted in the absence of the pump beam [23]. The theoretical scheme here proposed permits a full quantitative analysis of gain versus the input pump power. An increase of less than a factor of 1/3 in the polariton splitting produces an increase of the maximum achievable gain at $T = 10$ K [Fig. 3(a)] of more than 1 order of magnitude [Fig. 3(a)] in close agreement with experimental results [7]. The power dependence of gain (Fig. 3) shows an almost exponential growth and then saturates

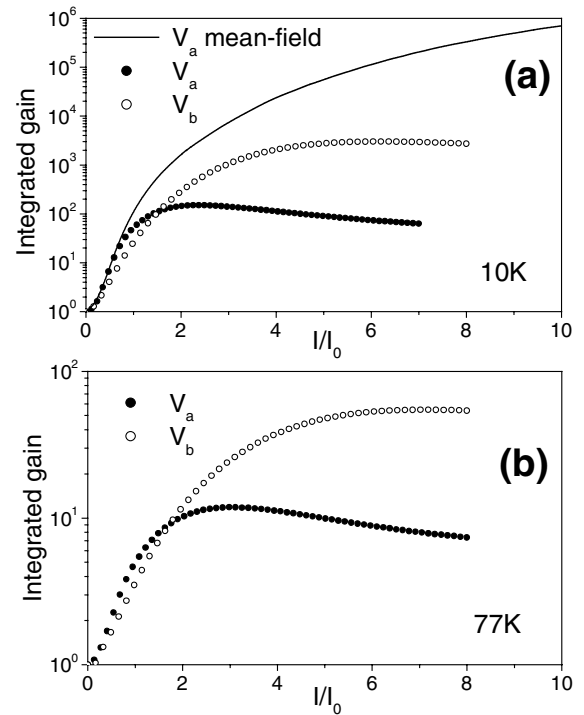


FIG. 3. Power dependence of the integrated gain calculated for two GaAlAs based samples with splitting $2V_a = 10.6$ meV and $2V_b = 15$ meV. $I_0 = 10^{13}$ photons per cm^2 per pulse, corresponding to 105 photons in a Bohr disk (πa_0^2) per pulse. The exciting light pulses have 250 fs duration. Material parameters of the two samples are coincident except for N_{eff} . The decay rate of cavity photons through the mirrors $\gamma_c = 0.25$ meV is that of GaAlAs structures considered in Ref. [7]. The 7 nm wide QWs have a binding energy $E_b = 13.5$ meV. Exciton homogeneous broadenings have been extracted from polariton linewidth measurements [22], $2\gamma_x = 0.6$ meV at $T = 10$ K (a), and $2\gamma_x = 1.4$ meV at $T = 77$ K (b). The gain curves have been obtained fully optimizing incidence angles, central frequencies of the input light beams and cavity mode energy.

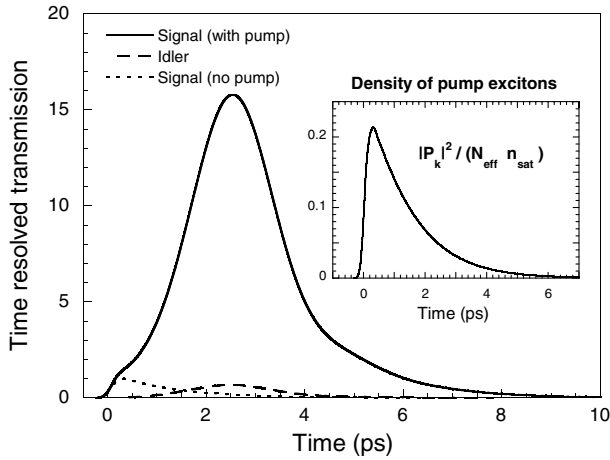


FIG. 4. Time-resolved transmission of the probe (with and without the presence of the pump beam) and additional emission (appearing in the presence of the pump beam) along the direction of the idler mode ($\theta \approx 47^\circ$), showing the ultrafast dynamics of the parametric amplification. The sample is V_b , the temperature is 10 K, and the pump intensity is $I = I_0$. The inset displays the time behavior of the pump-exciton density.

at high powers. The saturation of gain is mainly determined by the nonlinear absorption that is most relevant for the idler beam [according to Eq. (4) the idler nonlinear absorption is determined by $\text{Im}\mathcal{F}(\omega_{2k} + \omega_k)$]. We observe that the increase of the idler nonlinear absorbance is highly superlinear because the increase of the pump power produces both a direct increase of the exciton density and an increase of $\text{Im}\mathcal{F}(\omega_{2k} + \omega_k)$ as a consequence of the blueshift of the polariton-pair resonance $\omega_{2k} + \omega_k$ induced by $\text{Re}\mathcal{F}$. We observe that mean-field calculations [also reported in Fig. 3(a)] overestimate the experimentally observed gain of more than 4 orders of magnitude, neither are able to reproduce the strong dependence of gain on the exciton-photon coupling V , that is a direct consequence of the inner dynamics of XX collisions. Such a dramatic failure of the mean-field theory (at pretty high densities) is surprising and clearly shows the technological relevance that many-body correlations and their optical control may have in the future development of semiconductor all-optical devices. Figure 3(b) displaying results for $T = 77$ K, shows how the increase of the exciton-photon coupling favours high-temperature operation. At $T = 77$ K the sample V_a is still able to sustain significant gain about 6 times larger than that of sample V_b . The apparent discrepancy of the threshold values reported in Fig. 3 with the corresponding measurements [7] is due to a different definition of gain. Calculating the threshold values according to Ref. [7], we obtain $I = 0.8I_0$ (V_a) and $I = 0.96I_0$ (V_b) at $T = 77$ K, corresponding to a density of pump excitons $\max[|P_k|^2]/(N_{\text{eff}} n_{\text{sat}}) \approx 0.18$.

The low-temperature time dynamics of the parametric amplification is displayed in Fig. 4. The appearance of an additional light beam in a direction allowing momentum

conservation (idler mode), also observed experimentally [1] confirms the coherent nature of this scattering process involving pairs of polariton waves. The smallness of emission in the idler direction (as compared to the amplified signal) is a consequence of the low photon content of polaritons at high angles.

The results here presented explain why the polariton parametric amplifier works despite in exciton systems losses increase with the excitation density owing to decoherence produced by collisions between excitons. The results here shown demonstrate that exciton-exciton collisions in semiconductors can be controlled and engineered. Moreover, they give precise indications to produce almost decoherence-free collisions for the realization of all-optical microscopic switches and amplifiers. The availability of decoherence-free collisions appears crucial for the quantum control and manipulation of the polariton wave function inside the cavity and for the realization of microscopic sources of nonclassical light.

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