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Polariton-polariton scattering in semiconductor microcavities: Distinctive features and similarities to the three-dimensional case

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Polariton-polariton scattering has been measured in a GaAs-based microcavity by a c.w. polarized photoluminescence technique using circularly and elliptically polarized resonant excitation into the lower polariton (LP) branch. The scattering has been found to be strongly enhanced when the singlet biexciton state is allowed as an intermediate one, i.e., under elliptically polarized excitation. An exponential growth of $k=0$ LP emission has been achieved at high excitation densities for the major spin population leading to the emission polarization degree higher than that of the excitation. The effect indicates the scattering stimulation appearing due to the bosonic nature of polaritons when the high filling of $k=0$ LP states is reached.

Semiconductor microcavities (MCs) with embedded quantum wells represent a very interesting object for the investigation of light-matter interactions.[1–3] In such a system a strong two-dimensional (2D) confinement of light makes accessible very high densities of the photonic field which may result in a different type of nonlinear effects.[4,5] Another intriguing property is connected to the boson nature of the mixed exciton-photon states in MCs, 2D polaritons, characterized by an extremely small ($<10^{-4}m_0$, m_0 being the free electron mass) in-plane effective mass. The low density of polariton states makes possible the high filling at the LP band bottom to be achieved at relatively low densities when the influence of the fermion nature of electron and hole in exciton compound is yet negligible. In this case, the bosonic nature of polaritons can favor stimulated scattering[5,6] and allow Bose condensation. Experimental observation of these, presumably, nonlinear effects in the strong coupling regime is usually prevented by a slow relaxation of photoexcited polaritons into the LP band bottom.[7,8] Only recently the stimulated character of polariton relaxation has been demonstrated using resonant excitation into the lower or upper polariton branch.[9–11]

In the present paper we investigate the LP emission under conditions of the c.w. resonant excitation into the LP branch below the free exciton energy and thus we avoid any strong filling of free exciton states. Here we are interested in the study of peculiarities of polariton-polariton scattering in 2D case and the similarities of these processes to their 3D analoge in bulk semiconductors. The polariton-polariton scattering in bulk semiconductors was studied intensively in late 1970s and referred to as hyper-Raman scattering.[12] In general, the mechanism can be understood as four-wave mixing occurring with conservation of the total energy and wave number:

$$2E(\mathbf{k}_l)=E(\mathbf{k}_1)+E(\mathbf{k}_2), \quad 2\mathbf{k}_l=\mathbf{k}_1+\mathbf{k}_2 \tag{1}$$

where $\mathbf{k}_l$ [$E(\mathbf{k}_l)$] is the wave number (energy) of two polaritons in the initial state and $\mathbf{k}_{1,2}$ [$E(\mathbf{k}_{1,2})$] corresponds to two final states. The process has been shown to be squared in excitation power and highly enhanced when the biexciton state is included as an intermediate state in the scattering.[12]

Here we demonstrate that the LP-LP scattering in MCs reproduces the main features of the hyper-Raman scattering in bulk semiconductors following from the conservation laws and the resonance between the energies of the singlet biexciton state and $2E(\mathbf{k}_l)$. On the other hand, it has revealed striking peculiarities directly connected to the reduced dimensionality of cavity polaritons. First, we observe an exponential increase of the $k=0$ emission at relatively low powers of resonant excitation close to the point of inflection of the LP dispersion curve, which clearly indicates the stimulated character of the scattering. This occurs in a very similar way to the phenomenon observed by Savvidis *et al.* in pump-probe measurements[10] and theoretically described by Ciuti *et al.*[13] However, contrary to phenomena observed in pump-probe measurements,[10] where the additional weak probe was used to stimulate scattering of two pump polaritons, in our case the effect can be described as a self-stimulated process. Unlike the 3D case, the high filling of the 2D LP states at $k\approx 0$ becomes possible due to the *finite* LP energy $E(k\rightarrow 0)$ resulting in a qualitative change of the dispersion and density of the LP states at the band bottom. Second, in the case of excitation with circularly (σ) polarized light the exponential increase of $k=0$ population is observed only for major spin species. Furthermore, in the case of elliptically polarized excitation we observe a strong (more than two times) excess of the emission polarization degree over that of the exciting light. The effect has been shown to be connected to the difference of the final-state stimulation efficiencies of the LP-LP scattering originated from different filling of states with the total spin 1 and -1.

The sample studied is a high-quality single MC structure. The Bragg reflectors are composed of 17 (20) repeats of $\lambda/4$ $Al_{0.13}Ga_{0.87}As$/AlAs layers in the top (bottom) mirrors. The $3/2\lambda$ GaAs cavity contains six 10-nm-thick $In_{0.06}Ga_{0.94}As$/GaAs QWs. Measurements were carried out with the sample immersed in superfluid He. A c.w. Ti-Sapphire laser was used for resonant excitation of the sample. The photoluminescence (PL) signal was collected by a lens and a fiber mounted on a rotating rail. The signal was recorded by a 1-m double monochromator/CCD (charge coupled device) system. The experiments were carried out on several sample

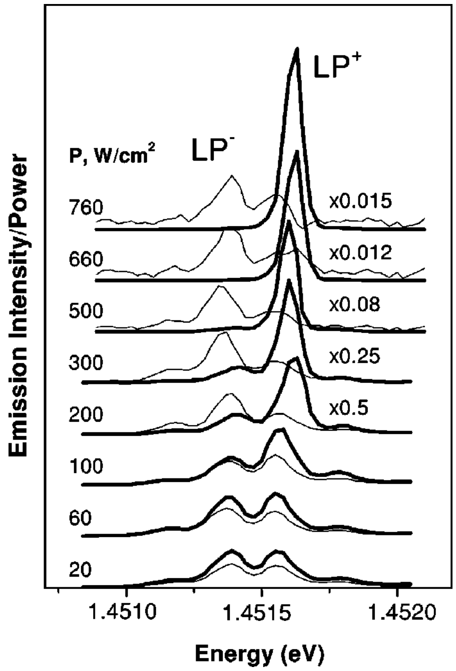

FIG. 1. Angle-resolved σ^+- and σ^--polarized LP emission spectra recorded at $\Phi=0°$ using the σ^+ polarized laser emission of various powers for resonant excitation into the LP branch at $\Phi_{\rm exc}=16°$ (all spectra are divided by P).

spots with Rabi splitting 6 meV and the detuning of uncoupled exciton and photon modes in the range of ± 1 meV.

Figure 1 displays angle-resolved σ^+- and σ^--polarized LP emission spectra recorded at $\Phi=0°$ using the σ^+-polarized laser emission of various powers for the resonant excitation into the LP branch at $\Phi_l=16°$. The spectra consist of a peak corresponding to the emission of LPs from the bottom of the branch. The fine structure of the peak is due to interference fringes within the sample (thickness $d \approx 0.5$ mm). At low P the energy and line shape of the emission peak in σ^+ (LP$^+$) and σ^- (LP$^-$) polarizations are similar. A larger intensity of the LP$^+$ emission ($I_{\rm LP^+}$) indicates that the time of spin and energy relaxation are comparable.[9] With increasing excitation density the line LP$^+$ becomes dominant and drastically narrows down to 0.075 meV, indicating the qualitative change in the LP relaxation.

The integrated intensities of the spectra in Fig. 1 are presented in Fig. 2. Figure 2 shows that $I_{\rm LP^+}$ and $I_{\rm LP^-}$ (squares and circles, respectively) depend linearly on P at $P \leqslant 100$ W/cm^2. Angle-resolved PL measurements show that in this range of P both $I_{\rm LP^+}(k)$ and $I_{\rm LP^-}(k)$ are symmetrical around $k=0$. The emission increases with Φ for $|\Phi|<\Phi_l$, which is consistent with the previous observation of the bottleneck in LP relaxation.[7,8]

At $P>100$ W/cm^2 the dependence $I_{\rm LP^+}(P)$ transforms into a squared one whereas $I_{\rm LP^-}(P)$ remains linear. Angle-resolved PL measurements have shown that in this regime a superlinear increase of the LP$^+$ line intensity occurs in a wide range of positive k, up to $k \approx 2k_l$.[14] As a result the LP$^+$ emission at $\Phi \geqslant 0$ is much stronger than that at $\Phi<0$. These changes point out the appearance of a new effective channel, namely, direct scattering of two LPs with momentum k_l into the states with momenta $k_l \pm \Delta k$. This four-wave-mixing process becomes highly effective when the exciting light is

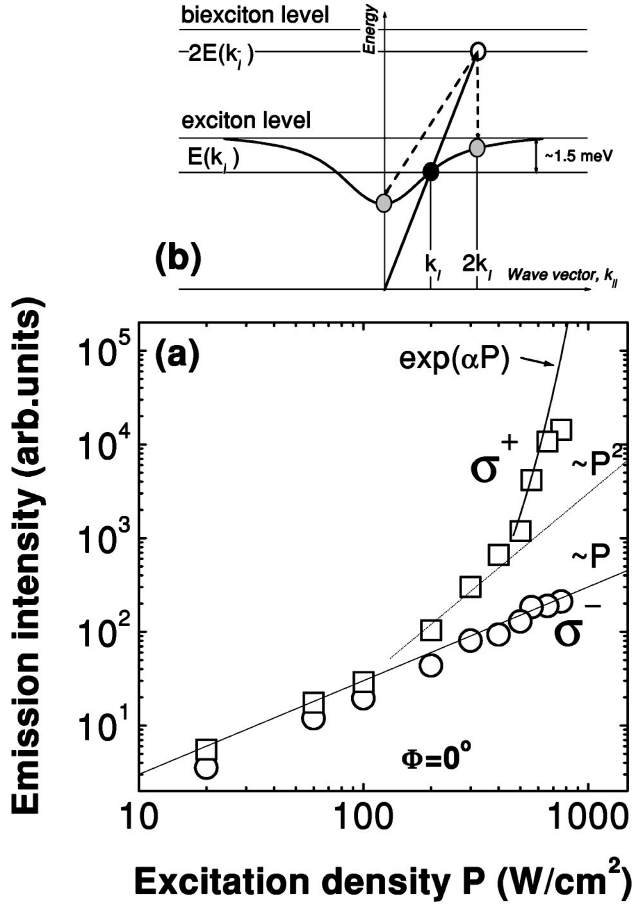

FIG. 2. (a) Dependence of LP emission intensity on excitation power for excitation with circularly (σ^+) polarized light (squares for LP$^+$ and circles for LP$^-$). (b) The diagram of the LP-LP scattering.

on resonance with the LP state[15] as shown in the diagram in Fig. 2 and can explain our experimental results, namely, (i) the square dependence on the excitation power, (ii) the mixing of the LP states with only positive k (the energy and momentum conservation laws [Eq. (1)] cannot be fulfilled simultaneously in our geometry for LP states with negative k), and finally, (iii) the contribution in the LP$^+$ line only (the four-wave mixing does not change the total LP spin). These features, based mainly on E and k conservation laws, are similar for 2D and 3D polaritons.

Based on the analogy with the scattering of bulk polaritons a strong enhancement of LP-LP scattering is expected when the two photon energy $2\hbar\omega(k_l)$ comes into resonance with the optically allowed biexciton transition. In our experiment $2\hbar\omega$ is only slightly, <1 meV, below the energy of the spin singlet biexciton state in the quantum well (see scheme in Fig. 2). Since the singlet biexciton state must be composed of two excitons of opposite spins, we can expect that the efficiency of LP-LP scattering should strongly depend on the polarization degree of laser excitation. This dependence has been measured with the use of elliptically polarized light excitation with the polarization degree $\rho_l=[I(\sigma^+)-I(\sigma^-)]/[I(\sigma^+)+I(\sigma^-)]$ varying from 1 to 0. Figure 3(a) displays the dependence of the overall $k=0$ LP emission, $I_{\rm LP}=I_{\rm LP^+}+I_{\rm LP^-}$, on ρ_l at $P=200$ W/cm^2 (solid squares). At this power the contribution from LP-LP scattering into the

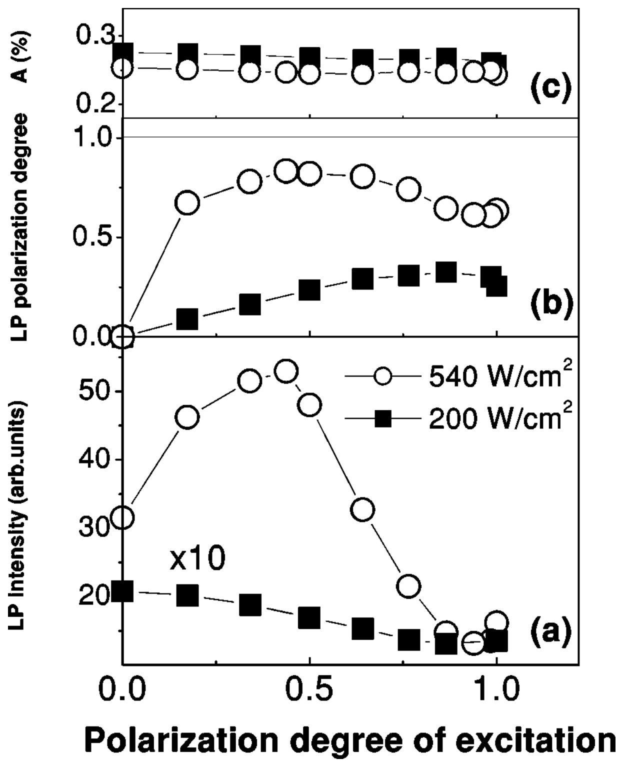

FIG. 3. The dependence $I_{\rm LP}$ (a) and $\rho_{\rm LP}$ (b) on the polarization degree of elliptically polarized exciting light for two excitation densities. (c) The dependence of light absorption on ρ_l. The data for $P=540$ W/cm^2 ($P=200$ W/cm^2) is shown by circles (squares).

relaxation of LP's is already significant. It is seen that the enhancement of $I_{\rm LP}$ reaches factor of 1.5 with decreasing ρ_l from 1 to 0. At the same time Fig. 3(c) shows that the absorption is practically constant in the whole range of ρ_l. The combination of these two observations constitutes clear evidence for the enhancement of the LP-LP scattering in MCs in conditions when the biexciton state becomes allowed as an intermediate one, similar to the behavior in bulk semiconductors.

The LP-LP scattering in MCs also reveals several distinctive features from the scattering of bulk LPs. First, the contribution from LP-LP scattering in MCs is observed at as small excitation density as ~ 150 W/cm^2. Such low threshold for LP-LP scattering originates from a huge magnification of electromagnetic field inside the MC. Another and much more striking feature originates from the reduced dimensionality of MC LPs. The finite LP energy at $k=0$ makes possible the accumulation of scattered 2D LPs at the band bottom, which, in its turn, may lead to stimulation of the LP-LP scattering process in accordance with their boson nature. Such stimulated scattering has been never observed in bulk semiconductors in the whole range of exciton densities up to 10^5 W/cm^2. In contrast, Figs. 1 and 2 show an abrupt exponential build up of the LP$^+$ emission in the MC excited with the σ^+ light already at $P\approx 450-500$ W/cm^2. This behavior cannot be described in terms of ordinary LP-LP scattering leading to squared power dependence. However, the LP-LP scattering as an origin for the exponential growth has been confirmed by PL measurements at large angles. We have found that in accordance with Eq. (1), this increase is followed by the corresponding nonlinear enhancement of the LP$^+$ emission at $k\approx 2k_l$. Thus we conclude that the LP-LP scattering in MCs acquires the stimulated character. This occurs already at $P\sim 500$ W/cm^2 due to the accumulation of LPs with $k\approx 0$. Thus unlike the pump-probe experiments, in our case the stimulation achieves an *intrinsic* character, i.e., it occurs without the additional probe beam. Figure 2 shows that the exponential dependence $I_{\rm LP}(P)$ saturates at $P\sim 700$ W/cm^2. The quantum efficiency of the LP emission in the angle $\Phi<2°$ also reaches its maximum at $P\sim 700$ W/cm^2 and does not change with a further increase of P. As shown in Figs. 1 and 2, no nonlinearity is observed in the emission of the minor LP spin component the filling factor of which remains very low in the whole range of P used. As a result, the LP polarization degree under conditions of stimulated scattering reaches as high a value as 97%.

The transition to stimulated LP-LP scattering results in several extraordinary features in the polarized spectra of the 2D LP emission under the elliptically polarized excitation. Note that the variation of ρ_l does not result in any marked change in the MC absorption [Fig. 3(c)] which implies that all surprising modifications in $I_{\rm LP}$ and polarization degree of LP emission, $\rho_{\rm LP}$, observed with changing ρ_l are connected with peculiarities in the LP scattering. First, Fig. 3(a) and 3(b) demonstrate nonmonotonic dependence of $I_{\rm LP}$ and $\rho_{\rm LP}$ on the polarization degree of excitation ρ_l for $P>250$ W/cm^2. It is seen that at $P=540$ W/cm^2 the increase in $I_{\rm LP}$ is ~ 3 times higher than that at $P=200$ W/cm^2 [Fig. 3(a)], and, in addition, the maximum of $I_{\rm LP}$ is shifted from 0 to $\rho_l\approx 0.4$. The initial increase in $I_{\rm LP}(\rho_l)$ with exciting light depolarization can be attributed to the appearance of the relaxation channel via intermediate biexciton state as described above. However this effect does not explain the decrease in $I_{\rm LP}$ at $\rho_l<0.3$ observed in the whole range of excitation densities $P>250$ W/cm^2.

Furthermore, the nonmonotonic dependence of $I_{\rm LP}(\rho_l)$ is accompanied by a nonmonotonic dependence of $\rho_{\rm LP}(\rho_l)$. In particular, Fig. 3(b) shows that $\rho_{\rm LP}$ at $P=540$ W/cm^2 increases rather than drops with the depolarization of the exciting light in a wide range of $\rho_l>0.34$ so that $\rho_{\rm LP}$ becomes *even larger* than ρ_l: it reaches ≈ 0.77 at $\rho_l\approx 0.34$. This behavior is completely different from that at $P=200$ W/cm^2 when $\rho_{\rm LP}$ is always smaller than ρ_l. Figure 4(a) compares the dependence of $\rho_{\rm LP}(P)$ for circularly ($\rho_l=1$) and elliptically ($\rho_l=0.34$) polarized light.[16] It shows that in the latter case the polarization degree $\rho_{\rm LP}$ reaches its highest value at noticeably lower P. This behavior correlates with a highly enhanced total intensity $I_{\rm LP}$ in the range of $P=300-500$ W/cm^2 due to appearance of the scattering via the biexciton state.

Thus one can conclude that at high P the spin population of $k=0$ LP's unbalanced above some critical value results in a much faster scattering of the majority spin LP's. The only mechanism which can explain this phenomenon is the final state stimulation, which is in our case evidently easier achieved for dominating σ^+ species. Apparently, the enhancement of the LP-LP scattering with decreasing ρ_l occurring due to the opening of the LP-LP scattering via the singlet biexciton state shifts the threshold for transition into the stimulated regime to lower excitation powers. This explains

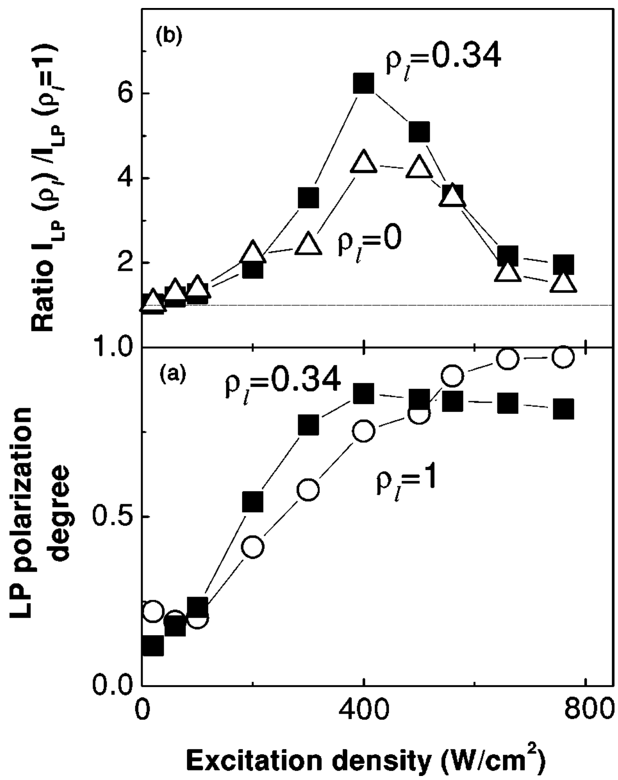

FIG. 4. (a) The dependence of the LP emission polarization degree on excitation density for the excitation with circularly ($\rho_l = 1$) and elliptically ($\rho_l = 0.34$) polarized light. (b) The power dependence of the ratio of the LP line intensity at excitation with elliptically ($\rho_l = 0.34$) and linearly ($\rho_l = 0$) polarized light to that at excitation with σ-polarized light.

the extraordinary increase in the polarization of LP line and the strong magnification of I_{LP} occurring with decreasing ρ_l in the range of $P = 300$–550 W/cm^2, i.e., just below the threshold to the stimulated scattering under the σ excitation.

Finally, in order to reveal the reason for the decrease of I_{LP} at $\rho_l < 0.34$ we plotted in Fig. 4(b) the power dependence of ratios $I_{\mathrm{LP}}(\rho_l = 0)/I_{\mathrm{LP}}(\rho_l = 1)$ and $I_{\mathrm{LP}}(\rho_l = 0.34)/I_{\mathrm{LP}}(\rho_l = 1)$. Although under linearly polarized excitation ($\rho_l = 0$) the emission is strongly enhanced compared to that measured using σ-polarized exciting light, the $I_L P(\rho_l = 0)$ is markedly smaller than $I_L P(\rho_l = 0.34)$ in the whole range of $P > 250$ W/cm^2. In this range the LP scattering gained the stimulated character and, hence, depends on the achieved filling of the LP states at the band bottom. Comparing the dependences I_{LP} and ρ_{LP} on ρ_l in Fig. 3 one finds that the beginning of the decrease of I_{LP} correlates with that of ρ_{LP}. The latter indicates that the LP's at the band bottom become distributed between two, σ^+ and σ^-, states, which results in the decreasing amplification factor and, hence, lower LP line intensity.

In conclusion, studies of the LP-LP scattering in the GaAs-based microcavity under c.w. excitation have revealed a strong contribution from four wave mixing of polariton states usually observed only at high excitations in pulsed measurements. The efficiency of LP-LP scattering increases when tuning from circularly to elliptically polarized excitation due to appearance of the relaxation channel including the singlet biexciton state as an intermediate transition. This behavior is similar to that of 3D polaritons in bulk semiconductors. However, in contrast to the 3D case, the strong stimulation of the scattering to the bottom of the LP branch has been achieved resulting in the exponential dependence of LP emission on the excitation power and in the pronounced nonlinearities in its polarization degree. The effect occurs in the strong coupling regime which justifies the application of the boson approach to the description of polaritons. The reduced dimensionality of LPs in MCs results in the finite LP energy at $k = 0$ and hence assists high filling of the states at the bottom of LP branch leading to stimulation of the LP-LP scattering.

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[14] We performed a detailed measurements of angle-dependence of the polarization and spectral properties of emission in a wide range of excitation powers of elliptically polarized excitation. This results are not presented here and to be published elsewhere.

[15] The LP dispersion curve is renormalized under conditions of strong excitations. However the renormalization is within 0.15 meV, i.e. negligible compared to Rabi splitting indicating the persistence of the strong coupling regime.

[16] The dependences in Fig. 3 are recorded from a sample spot different from that the results for which are presented in three other figures. Although the main features of observed phenomena are the same the enhancement factors for the I_{LP} and ρ_{LP} are slightly different.