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Nonlinearities in emission from the lower polariton branch of semiconductor microcavities

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Stimulated scattering of spin-polarized lower branch polaritons (LP) arising from their bosonic character is reported. The observations are achieved in a microcavity close to zero detuning between the uncoupled exciton and photon modes, under conditions of circularly polarized resonant excitation into the upper polariton branch. The key role played by the bosonic nature of the LP states is demonstrated by measurements of the degree of circular polarization of the emitted light P_{LP}. At low-excitation densities, P_{LP} is of the order of 30% in the whole range of wave vectors $k<2.10^4\ \mathrm{cm}^{-1}$. However, at higher excitation intensities, P_{LP} increases drastically, up to 80%, for LP's with $k\approx 0$, and is accompanied by the appearance of strong nonlinearities in the LP emission intensity whilst P remains unchanged for weakly coupled polaritons with $k>10^4\ \mathrm{cm}^{-1}$.
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Semiconductor microcavities (MC) modify the spectral and spatial distribution of photon fields and as a result change the nature and efficiency of light-matter interaction processes. The optical properties of MC's have attracted a high degree of interest ever since the first observation of the regime of strong light-exciton coupling in such structures.[1] This regime is realized in high finesse MC's with quantum wells (QW) embedded in the active layer when the coupling between the MC photon (C) and the QW exciton (X) modes exceeds their broadening. The resulting coupled exciton-photon system is described in terms of cavity polaritons by analogy to the excitonic polaritons of bulk materials.[2] Recent studies of nonlinear emission in MC's have been the subject of significant new interest and controversy.[3–6] Superlinear behavior in emission intensity has been observed at the energy of the photonlike mode, even in MC's with positive C-X detuning $\Delta=E_C-E_X$.[4,6] These observations were initially attributed to the effects of Bose statistics, and referred to as ''boser action.''[4] Such an explanation assumes excitons to be well-defined particles, a questionable assumption for the high-excitation densities used in Ref. 4. In particular, the superlinear growth of intensity in Ref. 4 was preceded by a strong decrease of the Rabi splitting indicating the suppression of the strong exciton-photon coupling. As a result alternative explanations of these effects were sought: A many body treatment of the coupled system of QW carriers interacting with the quantized light field was found to reproduce well the experimental observation within the framework of a transition from the strong to weak coupling regimes, rather than the occurrence of boser action.[6]

Recently Senellart and Bloch[7] reported the observation of superlinear behavior of the intensity of the photon-like polariton mode in an MC with large negative X-C detuning. These observations were achieved in the range of rather low-excitation powers when the Rabi splitting was unperturbed. The superlinearity was found to become weak with increasing excitonic character of LP's.

In this paper, we focus attention on the bosonic properties of the lower polariton (LP) mode in a III-V GaAs/AlAs MC with zero detuning when the states of the LP branch have large (50%) excitonic character. Quite recently Le Si Dang *et al.*[8] observed superlinear LP emission in the strong-coupling regime in II-VI MC's with $\Delta=0$. They suggested that one possible explanation for this behavior was stimulated bosonic scattering, favored by the relatively high-exciton binding energy in CdTe QW's.

We investigate here III-V MC's with $In_xGa_{1-x}As$ QW's but, in contrast to previous studies,[4,6,7] resonant circularly polarized excitation into the bottom of the upper polariton (UP) branch is employed. We find that the polariton system is markedly spin polarized, i.e., the spin relaxation time of the photoexcited polaritons exceeds their lifetime. With increasing excitation intensity we observe nonlinearities in the photoluminescence (PL) intensity which occur *only* for the majority polarization and occur below the onset of exciton bleaching. The study of the spin relaxation of photoexcited UP's as a function of excitation density enables us to obtain clear evidence for the bosonic character of the dense LP system in MC's with zero detuning.

The sample under investigation was a double MC structure grown by metal-organic vapor-phase epitaxy. The Bragg mirrors are composed of $\lambda/4$ $Al_{0.13}Ga_{0.87}As/AlAs$ layers of 12, 14.5, and 17.5 pair repeats, respectively, in the upper, intermediate and lower mirrors. Each GaAs cavity contains three 10-nm $In_{0.06}Ga_{0.94}As/AlAs$ QW's. Growth-related nonuniformities permit tuning of the photon mode energy, monitored by measuring the PL signal from different spots on the sample. The regions examined on the sample were determined by an Al mask deposited on the sample with small 20-μ diameter holes. The results reported here are obtained for the case when only one cavity mode has energy close to the exciton level. The other photon mode is located 10 meV higher than the lower UP mode and plays no significant role in the observed phenomena. The sample was maintained in a He cryostat at a temperature 5 K. A tunable Ti-Sapphire laser

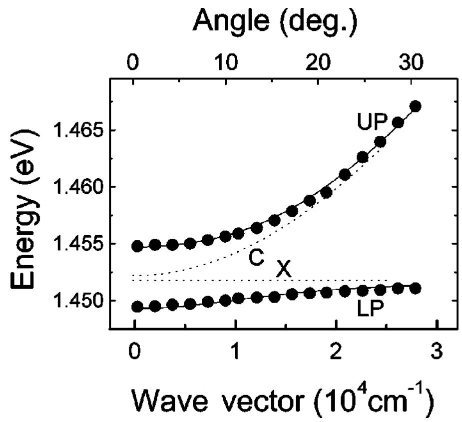

FIG. 1. Polariton dispersion obtained from low-excitation angle-resolved photoluminescence measurements.

was used for resonant excitation of the polariton states. The PL signal was dispersed by a double 1-m monochromator and detected by a nitrogen cooled charge coupled device (CCD) camera or GaAs photomultiplier. To avoid sample heating, chopped laser excitation was employed.

The experiments were carried out on several spots on the sample with Rabi splitting $\Omega = 5.4 \pm 0.3$ meV and X-C detuning $|\Delta| < 0.6$ meV. The values of Ω and Δ were determined from polariton dispersion curves measured by angle-resolved PL spectroscopy at low excitation intensity. A typical dispersion is displayed in Fig. 1 and shows that the X-C coupling is maximum at $\Phi \approx 0$ and becomes very small at $\Phi > 25°$.

Figure 2(a) displays σ^+ and σ^- polarized PL spectra for conditions of resonant excitation into the bottom of the UP branch ($\Phi = 3°$) with σ^+ polarized light. The PL spectra are recorded at $\Phi = 0$ (with an angular resolution of 0.5°) and correspond to the emission of polaritons with wavenumbers $k < 10^3$ cm^{-1}. The intensities of the spectra are normalized to the excitation power W. The dominant emission arises from the LP mode. At low W it is located at 1.449 eV in both

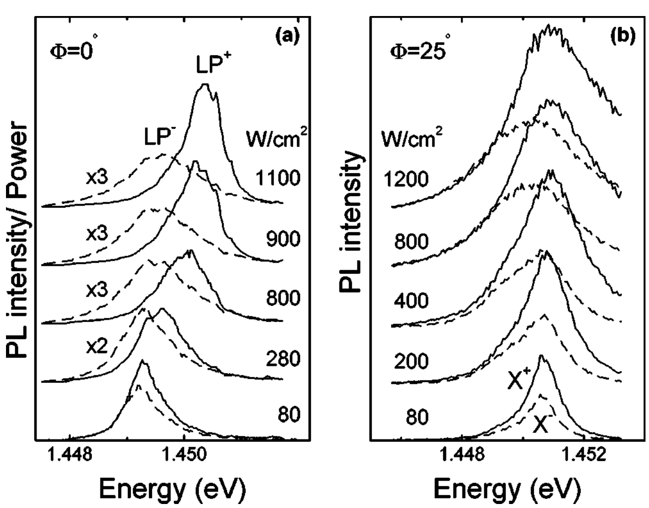

FIG. 2. σ^+ and σ^- polarized PL spectra at $\Phi = 0$ (a) and $\Phi = 25°$ (b) for various densities of σ^+-polarized resonant excitation. In (a) the PL intensity is normalized to the laser power and the numbers in the left column indicate the multiplication factors employed to display the σ^- spectra.

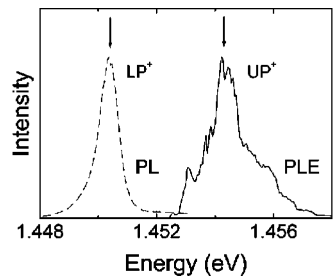

FIG. 3. PL and PLE spectra at $W = 1100$ W/cm^2. The arrows indicate the energies used for excitation and detection of the PL.

polarizations. The intensity of the peak in σ^+ polarization, LP$^+$, is greater than for σ^-, LP$^-$, implying that the spin-relaxation time is longer than the lifetime and that the photoexcited gas of polaritons is significantly spin polarized. The behavior of the LP$^+$ and LP$^-$ peaks with increasing excitation density is very different. The LP$^+$ peak shifts to higher energies by about 1.3 meV and exhibits a superlinear increase in intensity for $W > 600$ W/cm^2 and its width narrows slightly. By contrast, LP$^-$ increases slightly sublinearly in intensity, and displays a much smaller shift in energy but a stronger broadening.

The behavior of the $k = 0$ LP emission spectra is markedly different from those recorded at large k where the PL arises from the excitonlike polariton states. Reference to Fig. 1 shows that LPs acquire a dominant exciton character for $\Phi > 20-25° (k > 2.10^4$ cm^{-1}) when the X-C detuning exceeds 2Ω. PL spectra recorded at 25° are displayed in Fig. 2(b). They consist of a single line marked X. For small W the σ^+ to σ^- integrated intensity ratio for this line, I_X^+/I_X^- is very similar to that for the $k = 0$ LP peak in Fig. 2(a). However, in contrast to the strong increase of I_{LP}^+/I_{LP}^- with W, the ratio I_X^+/I_X^- changes only very little.

Figure 2(b) shows furthermore that the full width at half maximum of the line X becomes as great as 3.5 meV at high W. Such broadening indicates strong exciton-exciton interaction, as expected at high-exciton density, n_X. Indeed, a rough estimate[11] of n_X based on W and a typical exciton lifetime gives n_X of 1.2 to 3.6×10^{10} cm^{-2} for $W = 400 - 1200$ W/cm^2. The value of Ω can be determined directly from the $k \approx 0$ LP and UP energy gap. It is important to determine whether the sample is still in the strong coupling regime under conditions when the marked broadening of the X line occurs. To verify this the UP energy was determined from PL excitation (PLE) spectra of the $\Phi = 0$ LP$^+$ peak recorded with an angle of incidence of the laser beam of 3°. We observed a shift of 1 meV of the UP$^+$ mode to lower energy over the range of powers investigated. In order to obtain an unambiguous correspondence between the LP$^+$ and UP$^+$ mode energies with power, PL spectra were first recorded at fixed W for a set of excitation photon energies. Then the PLE spectrum was recorded for detection at the PL maximum. Figure 3 displays PL and PLE spectra obtained for $W = 1100$ W/cm^2. The UP$^+$ line is well resolved in the PLE spectra up to $W = 1300$ W/cm^2, two times greater than the threshold W for the superlinear behavior in the LP$^+$ mode intensity. The transition to the weak-coupling regime takes place at $W > 1500 - 1800$ W/cm^2 when a very narrow lasing line appears in the σ^+ PL spectra and the UP$^+$ peak

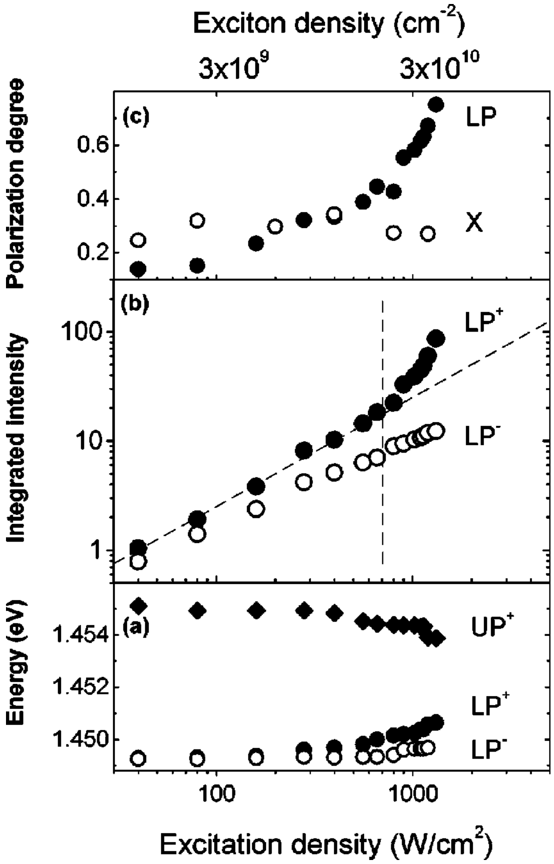

FIG. 4. (a) Energies of LP$^+$, LP$^-$, and UP$^+$ peaks vs the excitation density. (b) Integrated intensities of σ^+ and σ^- LP peaks at $k=0$. The dotted vertical line indicates the threshold power for the nonlinear behavior. The dash line corresponds to the linear dependence. (c) The degree of polarization for the $k=0$ (LP) and high $k=2.4\times10^4$ cm^{-1}(X) lower branch polariton states. The upper scale shows the estimated exciton density per QW (see text and Ref. 11).

becomes unresolvable in PLE. For different spots on the sample the lasing energy changes from 1.4510 to 1.4522 eV and is determined by the energy of the uncoupled photon mode (similar to earlier observations of Refs. 6 and 8). This regime is beyond the scope of this paper.

Figure 4(a) shows that the shift of the UP$^+$ and LP$^+$ modes with W is opposite in sign but similar in magnitude, indicating a reduction of the Rabi splitting for the σ^+ polaritons. Ω^+ decreases from 5.4 to 3 meV, with most of the decrease occurring for $W>800$ W/cm^{-2} due to exciton-exciton interactions. A simple estimate[13] shows that the exciton broadening of 3.5 meV itself is expected to lead to a relatively small, 0.7-0.8 meV, decrease of Ω. The stronger observed reduction of the LP$^+$-UP$^+$ splitting is thus probably due to a decrease of oscillator strength for majority, σ^+, excitons. The surprisingly large difference in the shifts of the LP$^+$ and LP$^-$ peak positions with W in Fig. 2(a) should be noted. Coulomb screening at high density is expected to cause a similar quenching of Rabi splitting for both polarizations. We believe there are two likely reasons for this behavior. Firstly, phase space filling and hence the corresponding change of the LP energy will be greater for X^+ than X^- and secondly stronger optical transitions from exciton to singlet biexciton states for X^+ than X^- are likely. Although detailed calculations are necessary for quantitative conclusions to be reached, we emphasize that the very small change in the LP$^-$ energy is strong evidence that the exciton system is represented by well defined quasi-particles in the whole range of $W<1300$ W/cm^2. We thus conclude that our experimental data show that the observed nonlinearities in the LP^+ emission intensity take place in the strong coupling regime.

Figure 4(b) shows that I^+_{LP} displays a strong superlinear, greater than quadratic variation for $W>600-700$ W/cm^2. We interpret this behavior as arising from the polariton density at the bottom of the LP$^+$ branch becoming sufficiently large to cause stimulated scattering into these states due to their bosonic character.

Further evidence is provided from study of polariton depolarization during energy relaxation from the $k=0$ UP states to the $k=0$ LP states. It is seen from Fig. 1 that direct acoustic phonon scattering of photoexcited $k\approx0$ UP$^+$ polaritons to the bottom of the LP branch is not possible.[9,10] It is well established that the UPs scatter first mainly to the X states with large, $>10^5$ cm^{-1},k which then scatter to the LP states with smaller k.[10,12] The energy of acoustic phonons[9] effective in polariton scattering is 1–2 meV, close to the energy difference between the LP states with $k\approx2\times10^4$ cm^{-1} and $k<10^3$ cm^{-1} (c.f. Fig. 1). As a result the polarization degree of the LP emission at $\Phi=0, P_{LP}=(I^+_{LP}-I^-_{LP})/(I^+_{LP}+I^-_{LP})$, is expected to be smaller than that of LP's with $k\approx2.10^4$ cm^{-1},P_X. The experimental dependence of P_{LP} and P_X is displayed in Fig. 4(c). It is seen that during the scattering process the photoexcited σ^+ polaritons undergo marked depolarization. At low W the degree of polarization decreases to $P_X=0.3$ at $k\approx2.10^4$ cm^{-1}, i.e., at the bottom of the exciton band, and decreases further to $P_{LP}\approx0.25$ at the bottom of LP branch. However with increasing W the polarization ratio P_{LP} changes to 0.8 (corresponding to $I^+_{LP}\approx8I^-_{LP}$) and becomes much larger than P_X, which changes only very weakly. Such a drastic increase of P_{LP} indicates strongly the enhancement of scattering into LP$^+$ states relative to LP$^-$, and demonstrates clearly the appearance of new mechanisms favoring scattering into the LP$^+$ states. Such non-linear behavior, favoring strongly just one sense of circular polarization for $k\approx0$ polaritons cannot arise from the nature of the scattering mechanism alone disregarding stimulation effects: phonon scattering which must proceed via high-k exciton states which have only small polarization ratio is spin independent, whereas exciton-exciton scattering although likely to favor the photoexcited σ^+ polarization, should also lead to a highly polarized $k\approx2.10^4$ cm^{-1} population, which is not observed. An additional enhancement of the scattering into LP$^+$ with small k is most likely due to final state stimulation, arising from the Boson character of the polaritons when the filling of LP$^+$ states exceeds one. Such stimulation is expected to have a marked threshold character and to increase strongly with final state filling, as observed.

Thus, the polarization selective measurements that demonstrate the superlinearity of only the $k\approx0$ LP$^+$ line intensity provide strong evidence that the nonlinearities observed in the strong-coupling regime are connected to the coherence of dense bosonic LP states. We also emphasize that although the effects occur at high density they do not arise from stimulated emission of photons; lasing at the energy of the uncoupled photon mode is seen, but only at higher density still when the system moves into the weak-coupling regime.

In conclusion, by employing resonant σ^+ excitation into the upper polariton branch we have found long polariton spin-relaxation times in MC's with zero X-C mode detuning. Weak excitation with polarized light results in 30% polarization of lower branch polaritons with $k < 3.10^4\ \mathrm{cm}^{-1}$. With increasing W we have observed a superlinear increase in the emission from LP$^+$ states with $k < 10^4\ \mathrm{cm}^{-1}$. We have demonstrated that these phenomena occur in the strong coupling regime from a combination of PL and PLE measurements. No superlinearities occur in the PL from the bottom of the LP branch in σ^- polarization, nor for either polarization for LP states with $k > 10^4\ \mathrm{cm}^{-1}$. Such behavior provides strong evidence that the nonlinearities arise from stimulated scattering into spin-polarized states at the bottom of LP branch, the stimulation arising as a result of the bosonic character of the polariton states. We stress that the use of circularly polarized excitation and detection techniques play a key role in the success of this work; they enable the population of the majority spin species to be probed directly, whilst study of the minority species provides simultaneous information on non-stimulated scattering processes.

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[1] C. Weisbuch, N. Nishioka, A. Ishikawa, and Y. Arakawa, Phys. Rev. Lett. **69**, 3314 (1992).

[2] V. Savona *et al.*, Phys. Rev. B **49**, 8774 (1994); S. Jorda, *ibid.* **51**, 10 185 (1995); R. Houdre *et al.*, Phys. Rev. Lett. **73**, 2043 (1994).

[3] R. Houdre *et al.*, Phys. Rev. B **52**, 7810 (1995).

[4] S. Pau, H. Cao, J. Jacobson, G. Bjork, Y. Yamamoto, and A. Imamoglu, Phys. Rev. A **54**, 1789 (1996).

[5] H. Cao *et al.*, Phys. Rev. A **55**, 4632 (1997).

[6] M. Kira, F. Jahnke, S.W. Koch, J.D. Berger, D.V. Wick, T.R. Nelson, G. Khitrova, and H.M. Gibbs, Phys. Rev. Lett. **79**, 5170 (1997).

[7] P. Senellart and J. Bloch, Phys. Rev. Lett. **82**, 1233 (1999).

[8] Le Si Dang, D. Heger, R. Andre, B. Boenef, and R. Romestain, Phys. Rev. Lett. **81**, 3920 (1998).

[9] F. Tassone, C. Piermarocchi, V. Savona, A. Quattropani, and P. Schwendimann, Phys. Rev. B **56**, 7554 (1997).

[10] J. Bloch and J.Y. Marzin, Phys. Rev. B **56**, 2103 (1997).

[11] Exciton densities were estimated from the excitation power and an exciton lifetime of 0.2 ns over the whole range of excitation densities. This lifetime was taken from the decay time measured under above band-gap photoexcitation and may be slightly larger than the polariton lifetime under resonant excitation. As a result the estimated values of n_X are likely to be an upper limit for the exciton density in the QW's.

[12] J.J. Baumberg, A. Armitage, M.S. Skolnick, and J.S. Roberts, Phys. Rev. Lett. **81**, 661 (1998).

[13] S. Pau, G. Bjork, J. Jacobson, H. Cao, and Y. Yamamoto, Phys. Rev. A **51**, 14 437 (1995).