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Stimulated scattering and its dynamics in semiconductor microcavities at 80 K under nonresonant excitation conditions

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We have observed stimulated scattering in a II-VI microcavity at 80 K under *nonresonant excitation conditions*. This stimulated scattering manifests itself through probe amplification in a pump-probe reflectivity experiment and through a highly nonlinear emission at an energy corresponding to a $k_{\parallel} \neq 0$ state on the lower polariton branch. We were able to follow the onset and decline of stimulated scattering by measuring the temporal evolution of the probe amplification.

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In semiconductor microcavities embedding quantum wells, mixed polariton states arise as a result of the strong coupling between the cavity photon modes and the quantum well excitons.[1] The strong mixing between photons and excitons drastically modifies the in-plane dispersion relations. Thus, the density of states of the mixed exciton-photon states can be four to five orders of magnitude smaller than that of excitons. In addition, due to the bosonic nature of polaritons, their scattering rate is proportional to $1+N$, N being the occupancy of the final state, where 1 corresponds to spontaneous and N to stimulated scattering. For N close to unity, the stimulated process dominates giving rise to a strongly nonlinear increase of the final state population and has been predicted to lead to a buildup of a coherent polariton population and polariton lasing (boser).[2] Because of the low polariton density of states, stimulated scattering effects can be expected for densities well below the exciton saturation density.

Indeed, recently several groups have reported evidence of stimulated scattering both in II-VI and III-V microcavities.[3–9,11,12] A strongly nonlinear dependence of the microcavity photoluminescence under nonresonant excitation was attributed to stimulated scattering.[4,7,8] For resonant excitation close to the point of inflection of the lower polariton dispersion curve, strong stimulated scattering was observed leading to $k_{\parallel}=0$ and high-$k_{\parallel}$ polariton populations.[6,9] This process was either self-stimulated by polariton scattering leading to a final state occupancy at $k_{\parallel}=0$ close to unity that initiates stimulated scattering[9] or stimulated by the presence of a probe injecting polaritons at the final state $k_{\parallel}=0$.[6] In the case of the pump-probe experiments,[6] a probe amplification of nearly two orders of magnitude was observed. More precisely, it is the presence of the polaritons injected by the probe that provokes stimulated scattering which leads to an inrease of the $k_{\parallel}=0$ polariton population by two orders of magnitude. The intensity of a $k_{\parallel}=0$ polariton emission is then also increased by the same amount.

In the present work, we have observed amplification of a probe pulse due to stimulated scattering following a *nonresonant excitation*. In contrast to resonant excitation that necessitates a spectrally well-defined pump pulse and, as a consequence, cannot provide good temporal resolution, the pump pulse in our case of nonresonant excitation was a femtosecond pulse. We are thus able to resolve the dynamics of the buildup of a polariton occupancy close to unity, the initiation and the decay of the stimulated scattering process. Furthermore, the stimulated scattering could be observed for a temperature as high as 80 K as opposed to all previous observations which required temperatures of 15 K or less. The final state associated to the stimulated scattering was not the $k_{\parallel}=0$ state but a higher-lying state and approaches the $k_{\parallel}=0$ state for low temperatures. We believe this state corresponds to the maximum polariton occupancy along the lower polariton dispersion branch.

The semiconductor microcavity used in this work was grown by molecular beam epitaxy on a $Cd_{0.88}Zn_{0.12}Te$ substrate. It consists of a $Cd_{0.4}Mg_{0.6}Te$ 3λ cavity containing 24 5-nm CdTe quantum wells (six groups of four quantum wells located at the maxima of the electric field distribution in the cavity). The top and bottom cavity mirrors are distributed Bragg reflectors made of 17.5 and 23 pairs of $Cd_{0.75}Mn_{0.25}Te/Cd_{0.4}Mg_{0.6}Te$ $\lambda/4$ layers, respectively. Due to the large exciton binding energy, the strong coupling regime persists up to room temperature. The Rabi splitting at 80 K was 26.4 meV.

Photoluminescence and pump-probe reflectivity measurements were performed using a Ti:sapphire femtosecond oscillator followed by a regenerative amplifier (200 kHz; Coherent RegA) and an optical parametric amplifier. The pump excitation pulse was nonresonant at 1.917 eV with a duration of 150 fs and was focused down to 120 μm. Both pump and probe arrived at normal incidence on the microcavity. The experiments presented below were performed for a detuning of -3 meV, that is an effective detuning (ratio of detuning to Rabi splitting) of -0.11. Similar results were obtained for a detuning of -13 meV. Great care was taken to probe only a homogeneously excited area of the sample. Therefore, only the central 20 μm of the photoluminescence and of the

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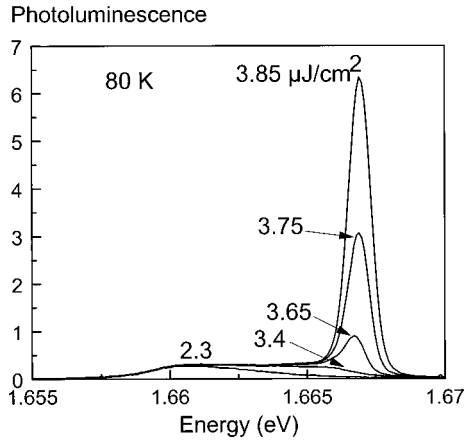

FIG. 1. Photoluminescence spectra normalized for the excitation density. A strong nonlinearity is observed above a threshold of 3.5 μJ/cm^2.

probe in the pump-probe experiments, respectively, were selected with a pinhole at an intermediate image and sent into the spectrometer and CCD detector. The probe differential reflectivity, $\Delta R/R$, due to the presence of the pump was measured at an 8-Hz rate in order to suppress the noise due to low-frequency laser fluctuations. The reflectivity spectra in the presence of the pump were then reconstructed from the differential reflectivity and the reflectivity in the absence of the pump.

We first performed photoluminescence experiments involving the pump only. A highly nonlinear emission was observed in this sample at $T=80$ K above an excitation-density threshold value of 3.5 μJ/cm^2 (see Fig. 1). This nonlinear emission is similar to that reported in Refs. 3 and 7 for lower temperatures (4 and 15 K) (Ref. 13) and also shows a spectral narrowing. In the latter case, the dependence of the nonlinear emission threshold on the number of quantum wells and on the detuning between the exciton and photon mode lead to the attribution of the nonlinear emission to stimulated exciton-exciton scattering. In our case, it is interesting to note that the nonlinear emission is shifted 7 meV to higher energies with respect to the linear emission which appears at the same energy as the lower polariton reflectivity signature. Only about 1 meV of this shift is due to a blueshift of the lower polariton at higher excitation densities (as we will see in the reflectivity spectra in Fig. 2) and we attribute the remaining 6 meV to a participation of polaritons on the lower polariton branch with $k_{\|}\neq 0$. This observation is not surprising given the results of Refs. 10,14 where an accumulation of polaritons for $k_{\|}\neq 0$ was reported. A shift, though of smaller value, was also observed in experiments done at low temperature[3,4,6,7] and there also it could not be entirely explained by the shift of the lower polariton under excitation. Pump-probe reflectivity spectra taken in the same conditions as the nonlinear emission show that this emission appears while the microcavity is still in the strong coupling regime as was the case in Ref. 7.

Selected reflectivity spectra are shown in Fig. 2 for different pump-probe delays and for a pump excitation density of 3.75 μJ/cm^2 just above the nonlinear emission threshold in-

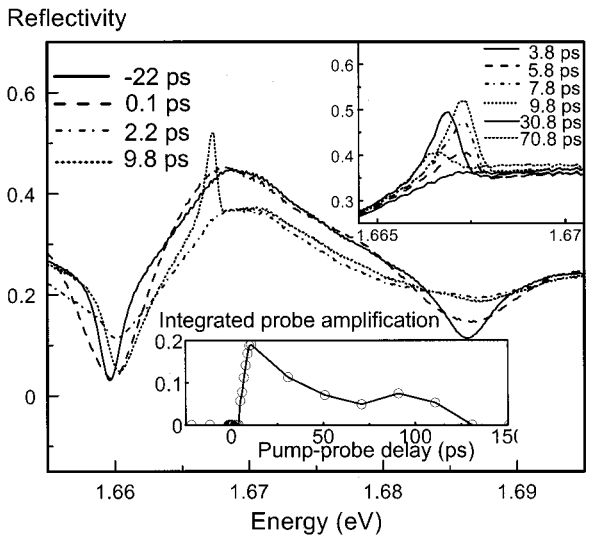

FIG. 2. Selected reflectivity spectra at $T=80$ K for different pump-probe delays and for a pump excitation density of 3.75 μJ/cm^2. The upper inset is a blow-up of the spectral region where the probe amplification is observed. The lower inset shows the spectrally integrated probe amplification in arbitrary units as a function of pump-probe delay.

jecting about 10^{11} cm^{-2} electron-hole pairs in the quantum-well continuum. The pump-probe delay values were obtained from the period of the coherence oscillations observed at negative delays.[15] The solid curve corresponds to the unperturbed sample (before the pump arrival). The dashed curve (0.1 ps) and the dashed-dotted curve (2.2 ps) taken during and after the pump pulse, respectively, show an increasing broadening of both the upper and lower polariton peaks. Several picoseconds after the end of the pump, the broadening of the polariton peaks has partly recovered and a small blueshift of both polaritons is clearly observed. Furthermore, an amplification of the probe shows up on the high-energy side of the lower polariton peak at the same energy as the nonlinear emission observed in the absence of the probe (see Fig. 1). The dynamics of this amplification is presented in the upper inset of Fig. 2.

It should be noted that this amplification signal cannot be attributed to the photoluminescence of the microcavity: The latter is due to the presence of the pump only and does not depend on pump-probe delay. The small contribution of the photoluminescence has been subtracted from the differential reflectivity spectra at large negative delays before converting them into reflectivity spectra. The probe is amplified by less than a factor of 2 but we expect much larger values for a detection angle corresponding to the $k_{\|}$ of the participating polaritons. Indeed, our maximum collection angle of 8° is much smaller than the external angle of 23° ($k_{\|}=3.4\times 10^4$ cm^{-1}) corresponding to the observed polariton energy. The detected photons are thus only a small fraction of the emitted photons and we believe that they are detected due to elastic scattering at the sample surface or interfaces.

We attribute this probe amplification to stimulated exciton-exciton scattering. The proposed process is the following (see schematic representation in Fig. 3): The pump

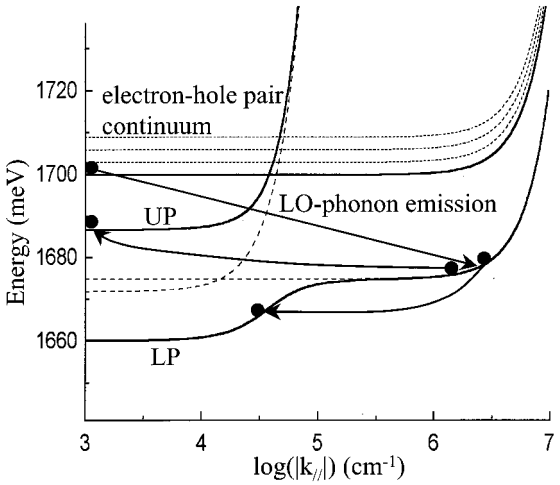

FIG. 3. Dispersion relation of the upper (UP) and lower (LP) polariton branches (solid lines). The dashed lines show the exciton and photon dispersion in the absence of coupling, the short-dashed lines show the dispersion of some electron-hole-pair continuum states, while the arrows show the process proposed to explain the probe amplification. Note that in the process shown the wave vector is conserved. The fact that the wavevector change appears to be different for the two polaritons participating in the scattering process is due to the logarithmic wave vector scale.

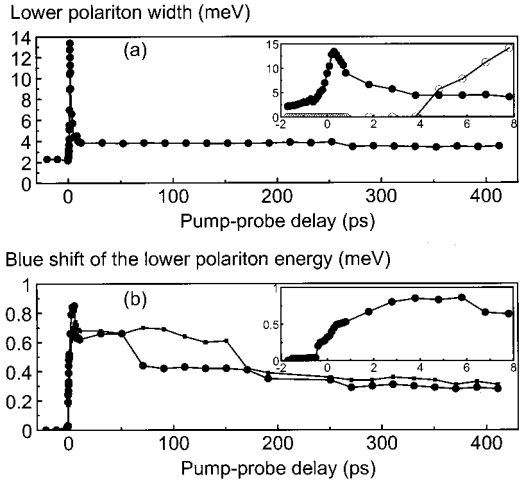

FIG. 4. Lower polariton width (a) and blueshift of the lower polariton energy (b) as a function of pump-probe delay obtained from Lorentzian fits of the reflectivity spectra for an excitation density of 3.75 μJ/cm^2 (full circles). The error bar is ± 0.1 meV. The insets are a blow-up of the short pump-probe delay range. The inset of (a) also shows a blow-up of the spectrally integrated probe amplification shown in Fig. 2 (open circles). The squares in (b) correspond to an absorbed pump excitation density of 1.1 μJ/cm^2.

pulse creates electron-hole pairs in the quantum-well continuum. These electron-hole pairs form excitons via LO-phonon emission.[16] Subsequently, two excitons with $k_\parallel$ of opposite sign scatter resulting in an upper and a lower polariton. When the population of the lower polariton state corresponding to the final state of this exciton-exciton scattering process approaches unity, stimulated scattering sets in causing a strongly nonlinear increase of the lower polariton population. This stimulated exciton-exciton scattering continues as long as there is a sufficient initial population in the exciton reservoir. When the initial population for this scattering process declines, the lower polariton population also drops below unity due to the emission process and stimulated scattering ceases. In the presence of the probe, the scattering rate is proportional to $1+N_{\mathrm{pump}}+N_{\mathrm{probe}}$, where N_{pump} is the lower polariton occupation factor obtained after LO-phonon emission and scattering from the electron-hole pairs injected by the pump and N_{probe} the lower polariton occupation factor resonantly injected by the probe.

Under stimulated scattering conditions (N_{pump} close to and above unity), the presence of N_{probe} leads to an increase in the scattering rate which translates into an increase of the lower polariton population and thus an amplification of the probe. The probe being weak, the main part of the population giving rise to the stimulated emission is built up due to the relaxation of the electron-hole pairs injected by the pump. This is confirmed by the fact that for pump intensities below threshold (data available for 2.2 μJ/cm^2 and below) no probe amplification is observed. We therefore believe that the presence of the probe allows to visualize the dynamics of the stimulated scattering process without substantially altering it. As a consequence, the spectrally integrated probe amplification shown in the lower inset of Fig. 2 reflects this dynamics. Stimulated scattering starts at about 4 ps, shows a maximum at 10 ps, and continues upto approximately 130 ps. There is a small spectral shift (1 meV) of the probe amplification as a function of pump-probe delay (see upper inset of Fig. 2). About half of it is related to a shift of the lower polariton peak. The rest of it probably translates a displacement of the stimulated scattering process towards smaller $k_\parallel$ wave vectors.

The above interpretation is corroborated by the behavior of the width and peak position of the lower polariton peak. It is depicted in Fig. 4 (full circles) as a function of pump-probe delay and was obtained from the full set of reflectivity spectra under the conditions of Fig. 2. The insets correspond to a blow-up of the short pump-probe delay range. The energy positions were obtained with Lorentzian fits of the polariton peaks. The lower polariton width at half maximum increases rapidly during the pump pulse from 2 to 14 meV. A rapid recovery of the width down to 4 meV in about 4 ps is followed by a slow recovery. The lower polariton energy exhibits a drastically different behavior: It continues to increase until about 4 ps after the end of the pump pulse [see inset of Fig. 4(b)].

These results are interpreted as follows: As soon as the pump pulse creates electron-hole pairs in the continuum, these electron-hole pairs effectively scatter the lower and upper polaritons at $k=0$ by interacting with the polariton exciton components thus increasing their broadening. When excitons are formed, the polariton broadening seen by the probe partly recovers because the scattering rate of polaritons with excitons is much lower than with electron-hole pairs since excitons are neutral particles. The exciton formation also has another consequence: Due to the hard-core repulsion between excitons,[17–19] the exciton energy, and subsequently

the energy of both polaritons, shift to the blue as is observed in Figs. 2 and 4(b). Indeed, the partial recovery of the polariton width is concomitant with the increase in polariton energy which leads us to attribute the 4-ps characteristic time related to both effects to the exciton formation time. This exciton formation time is shorter than the exciton formation times measured in GaAs quantum wells.[16] We attribute this observation to the fact that in these CdTe quantum wells the LO-phonon energy (21.4 meV) is very close to the exciton binding energy (25 meV) thus leading to very efficient exciton formation through emission of LO phonons with small wave vectors. [In GaAs quantum wells, the LO-phonon energy (36 meV) is much larger than the exciton binding energy of about 10 meV.] These two effects, rapid recovery of the lower polariton width and delayed increase of the lower polariton energy, and their dynamics are consistent with the onset of probe amplification at about 4 ps [see lower inset of Fig. 2 and its blow-up shown with open circles in the inset of Fig. 4(a)].

Furthermore, the blue shift of the lower polariton decreases following the decline of the probe amplification and thus the decline of the population in the exciton reservoir. In order to confirm the above interpretation, we also show in Fig. 4(b) the dynamics of the lower polariton energy (squares) for a pump excitation density below the threshold of the nonlinear emission observed in the absence of the probe ($1.1\ \mu J/cm^2$). In this case, no probe amplification takes place and the blue shift of the lower polariton reflecting the population in the exciton reservoir decreases more slowly than in the case above threshold where stimulated scattering empties the exciton reservoir.

In conclusion, our results demonstrate that stimulated scattering and probe amplification can be observed even under nonresonant excitation which is promising for potential applications. This is contrary to the predictions of Ref. 20, however, also earlier experimental results[10] have shown that the bottleneck effect calculated in Ref. 20 was overestimated. Furthermore, we have observed nonlinear emission and probe amplification under strong coupling conditions at temperatures as high as 80 K. The evolution of the probe amplification as a function of time indicates that the buildup of a lower polariton population close to unity from electron-hole pairs injected into the quantum-well continuum takes about 4 ps. Stimulated exciton-exciton scattering is then sustained upto about 130 ps.

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