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Stimulation of Polariton in Widebandgap Semiconductor Microcavities

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1. Introduction

Semiconductor microcavities are planar Fabry-Perot cavities of length comparable to the emission wavelength of the active medium, which is usually one or more quantum wells (QWs) situated at the antinodes of the cavity photon mode. These devices have attracted new physical interests since the observation of the strong-coupling regime by Weisbuch et al. [1]. This regime is realized when the coupling between the exciton confined in the QW and the photon mode confined in the cavity exceeds their own damping and lifetime. Experimentally the strong-coupling regime is most easily demonstrated by the anticrossing behavior exhibited by the exciton and the cavity mode when they are brought into resonance. The resulting coupled exciton-photon states are considered as (upper and lower) cavity polaritons, in analogy to bulk exciton polaritons [2].

Recently Imamoglu et al. [3] pointed out that, due to the bosonic nature of excitons, the exciton-phonon relaxation process could stimulate the generation of the lowest energy polariton state. According to these authors the stimulation condition is fulfilled when the exciton reservoir occupancy exceeds that of the phonons. This may require a high generation rate of excitons, which could result in their bleaching, and the subsequent breakdown of the strongcoupling regime. Previous studies of GaAs-based microcavities under high excitation conditions seem to support such sequence of events since all observed optical nonlinearities with threshold-like behaviors can be explained in terms of electron-hole correlations in the weakcoupling regime [4].

If exciton bleaching is the limiting factor for polariton stimulation, II-VI wide bandgap semiconductor microcavities appear to be better candidates because exciton Bohr radii are much smaller in these materials. In this paper we present a photoluminescence (PL) study of CdTe-based microcavities under high optical excitation densities at 4.2 K. Two distinct stimulation effects are observed with increasing excitation. The first one occurs in the strong exciton-photon coupling regime, implying stimulated generation of cavity polaritons. It confirms the early stimulation result observed in another CdTe-based microcavity by Bleuse et al. [5]. The second stimulation, obtained for much higher excitation, is the usual lasing action by population inversion of the electron-hole plasma in the weak-coupling regime.

2. Results and discussions

A series of four microcavities containing one, two, six or sixteen CdTe QWs are grown by molecular beam epitaxy on (001)-Cd_{0.88}Zn_{0.12}Te substrates. The QWs, 50 Å thick, are inserted in Cd_{0.80}Mn_{0.20}Te cavities. The top and bottom cavity mirrors are distributed Bragg reflectors made of thirteen and eighteen pairs of $\lambda/4$ Cd_{0.75}Mn_{0.25}Te / Cd_{0.40}Mg_{0.60}Te layers, respectively. The maximum reflectivity in the stop band is estimated to be about 0.95. PL is measured by exciting into the continuum of QW excitons (at about 1.8 eV, above the reflectivity stop band). The excitation source is a dye laser pumped by a frequency doubled Nd:YAG laser which delivers 5 ns pulses at a repetition rate of 10 Hz. Reflectivity and PL measurements are analyzed with a Jobin-Yvon THR-1000 monochromator equipped with a CCD detector.



Fig. 1 Optical measurements of the microcavity containing 16 QWs, at 4.2 K and for zero exciton-photon detuning. (a) Reflectivity spectrum. (b) PL spectra excited at 1.8 eV for a range of excitation densities. All spectra are normalized to the corresponding excitation densities. The A line is attributed to polariton stimulation in the strong exciton-photon coupling regime. (c) Same as in (b). The B line corresponds to lasing action of electron-hole plasma in the weak-coupling regime.

Fig. 1a shows the reflectivity spectrum of the microcavity containing sixteen QWs for detuning close to zero, i.e. when the cavity mode is nearly resonant with the 1s ground state of the heavy-hole exciton in QWs. The upper and lower polariton states are well resolved (FWHM $\approx 2.7 \text{ meV}$) at 1681.6 and 1658.7 meV, respectively. The measured Rabi splitting of about 23 meV is consistent with an exciton oscillator strength $f \approx 2.3 \times 10^{13} \text{ cm}^{-2}$ and an estimated effective number of quantum wells $n_{eff} \approx 14$. Figs.

1b and 1c show PL spectra of the same microcavity for zero detuning, as a function of the excitation density. Note that all PL spectra are normalized to the corresponding excitation densities. Thus a superlinear (sublinear) dependence of PL intensity on excitation should give an increase (decrease) of the normalized spectra.

As shown in Fig. 1b, the low excitation spectrum consists of a single line at 1659.7 meV, which can be assigned to the spontaneous emission from the lower polariton. Its width is about the same than in the reflectivity spectrum. No emission from the upper polariton is observed at low temperatures due to thermalization effects. The lower polariton emission increases linearly with excitations up to about 40 kW/cm² since its normalized PL remains unchanged. Then for a further increase of excitation a new line, labeled A, emerges from the high-energy shoulder of the lower polariton line. The sharpness of the line (FWHM ≈ 0.8 meV as compared to the bare (uncoupled) cavity FWHM \approx 4 meV) and its sudden appearance resemble to the onset of stimulated emission. More interesting is its spectral position at 1661.4 meV. It is well below the position of 1670 meV, which would be the energy of the uncoupled QW exciton and cavity mode for zero detuning. This clearly shows that the exciton-photon coupling is still in the strong regime, and hence the polariton description remains valid. A rough estimate of the exciton density at threshold would give 2.9x10¹⁰ cm⁻², which is about one order of magnitude smaller than the exciton saturation density in 50 Å CdTe QWs. Therefore we assign the A line to the stimulated generation of polaritons.

Fig. 1c shows PL spectra under higher excitation conditions. Starting from 40 kW/cm² the A emission line increases linearly with excitation up to about 150 kW/cm², without any change in its width and in its position. Then for higher excitations up to about 1 MW/cm² it broadens continuously and slightly shifts to the blue (by less than 1 meV). For still higher excitations another line, labeled B, appears on the high-energy shoulder of the A line. It shifts to the blue to the final position at 1670 meV that corresponds exactly to the bare cavity mode (that is also the bare QW exciton for zero detuning). Now excitons are completely ionized and the microcavity system is switched to the weak-coupling regime. Lasing at the cavity photon mode (B line, FWHM $\approx 2.5 \text{ meV} \approx \text{half}$ the bare cavity width) is observed for a threshold of about 10 MW/cm², which is two orders of magnitude higher than for the polariton stimulation.

It is found that polariton stimulation is most easily obtained in microcavities tuned to resonance with a large number of QWs. More precisely, the threshold roughly scales as the inverse of the number of QWs in the microcavity. This dependence is just opposite to that expected for the usual lasing action in edge-emitting QW lasers or Vertical-Cavity-Surface-Emitting-Lasers whose threshold always increases with the number of QWs. Fig. 2 shows how threshold for polariton stimulation varies with exciton-photon detuning in the microcavity containing sixteen QWs. It increases exponentially with negative detuning, except for a narrow range of detuning indicated by the arrow in the figure. In fact a closer examination of polariton dispersion in the QW plane reveals that exciton-phonon scattering should be accelerated in this range of detunings since it corresponds to an energy difference of one LO phonon (21.3 meV) between exciton states with large (in-plane) $k_{//}$ and the lower polariton state with $k_{//} = 0$. This finding gives strong support to the exciton-phonon relaxation mechanism suggested by Imamoglu et al. [3] for the stimulation of polaritons.



Fig. 2 Threshold of polariton stimulation versus detuning for the microcavity containing 16 QWs (see text). The dashed line is only guide for eyes.

3. Conclusion

In conclusion, it is unambiguously shown that polaritons can be stimulated at low temperatures in CdTe-based microcavities. This novel effect should be even more favored by stronger excitonic effects in wider bandgap semiconductors such as ZnSe or GaN. Its physical origin is not fully understood yet. Our preliminary results suggest that exciton-phonon scattering is involved in the stimulation process, as early suggested by Imamoglu et al. [3].

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