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Enhancement of spontaneous emission rates by three-dimensional photon confinement in Bragg microcavities

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Three-dimensionally confined Bragg microcavities exhibiting several fully quantized photon modes have been investigated by time-resolved photoluminescence spectroscopy. A strong decrease of the exciton lifetime with decreasing lateral dimension is observed and shown to be mainly due to coupling of the long-lived excitons with short-lived photon modes. We observe similar decay times of all photon modes for a fixed structure size which amount to approximately half of the corresponding exciton lifetimes. This behavior is explained by incoherent Auger-type scattering processes populating the photon modes on the low-energy side of the exciton line. [S0163-1829(97)51632-8]

Planar semiconductor microcavities with distributed Bragg reflectors have been extensively studied in recent years, both theoretically and experimentally, due to remarkable possibilities for controlling the exciton optical properties via the coupling with the cavity resonance photon mode. Rabi splitting in the cavity reflection spectra has been observed due to the polaritonlike coupling between the exciton and vertical photon mode of the resonator.[1] It has been anticipated that exciton-photon coupling could also cause significant shortening of the spontaneous exciton lifetime, which would be very important for device applications. Measuring the lifetime of the individual coupled exciton-photon states under resonant excitation, Sermage *et al.*[2] have demonstrated a significant reduction of the spontaneous emission decay rate at small detunings below 5 meV. Under nonresonant excitation, however, an exciton population with a broad distribution of in-plane wave vectors $k_{\|}$ is produced which radiates mainly into the leaky modes of the Bragg reflectors and into the continuum of horizontally waveguided modes of the planar microcavity.[3,4] Therefore, no significant influence of the exciton-photon coupling on the measured *effective* spontaneous emission lifetime is expected.[5,6] Experimentally, lifetime changes of a few ten percent are reported in Refs. 7–10.

Very recently, studies of semiconductor microcavities with three-dimensional optical confinement have been reported.[11,12] In these structures, vertical optical confinement by the epitaxially grown high-quality Bragg mirrors is combined with lateral optical confinement due to the large refractive index discontinuity at the etched sidewalls. Discrete optical eigenmodes with quantized $k_{\|}$ of several orders have been observed.

In the present paper, we investigate experimentally the photoluminescence kinetics in such structures, and demonstrate a strong shortening of the exciton lifetime by a factor of 6 when the lateral size is decreased from 100 to 1.7 μm. This decrease is mainly attributed to the coupling between the exciton and discrete photon modes in zero-dimensional (0D) microcavities. The emission decay times of the photon modes observed as far as 20 meV below the exciton line of the 0D microcavities are seen to be further reduced by approximately a factor of 2. They are controlled by the longer exciton decay time through incoherent Auger-type relaxation processes.

The stacking layers for the present three-dimensional microcavity structures have been grown by molecular-beam epitaxy. 17 (21) highly reflective AlAs/GaAs mirror pairs constituting highly reflective distributed Bragg reflectors (DBR's), are located below (above) a GaAs cavity with a dimension of one wavelength (250 nm) of the $In_xGa_{1-x}As$ quantum well (QW) emission. Three $In_{0.14}Ga_{0.86}As$ QW's (7 nm well width) separated by barriers with a thickness of 10 nm are placed at the center of the λ cavity, i.e., at the antinode position of the electric field. Arrays of quadratic microcavities with lateral sizes between 5 μm and 1 μm were obtained by electron-beam lithography and dry etching through the top mirror stack and the cavity.[13] From part of the planar microcavity sample the upper AlAs/GaAs mirror was removed by wet chemical etching in order to investigate the carrier dynamics without a resonator in otherwise identical structures (see inset of Fig. 1).

Time-resolved photoluminescence experiments were performed at 5 K using a mode-locked Ti:sapphire laser as an excitation source (1.5 ps pulses). The excitation energy at 1.468 eV selectively excites the $In_xGa_{1-x}As$ QW's only and was chosen above the stop band of the Bragg mirrors. The PL signal was dispersed by a $f=25$ cm monochromator and detected with a synchroscan streak camera. The temporal resolution of the setup amounts to about 10 ps.

Figure 1 shows the time-integrated spectra of 2.7 μm wide microcavities (full line) in comparison with data of 2.7 μm wide structures without the top Bragg mirror (dashed line) as illustrated by the inset. The excitation density of 1000 W cm^{-2} was kept well below the threshold for stimulated emission of the microcavity modes. For structures without cavity the excitation intensity was reduced in order to account for the reflection losses at the top Bragg mirror of

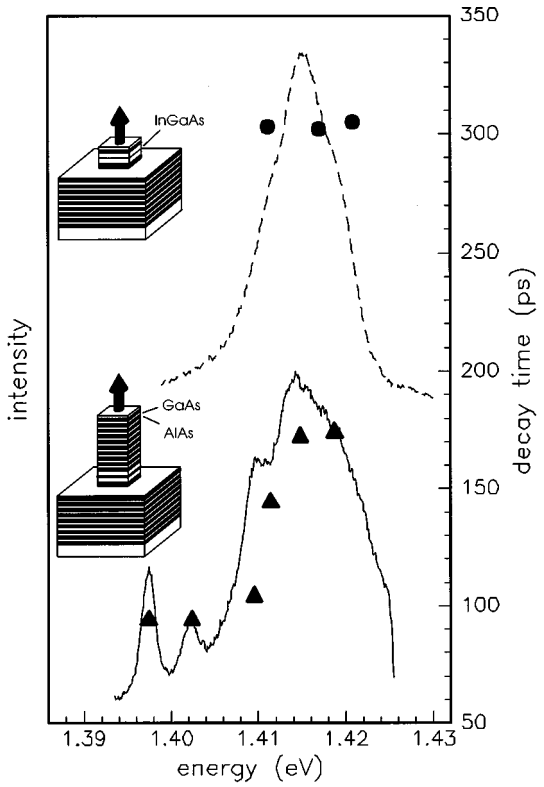

FIG. 1. Time-integrated spectra of three-dimensionally confined microresonators with lateral dimension of 2.7 μm (full line) and equally sized structures without the upper Bragg mirror (dashed line) as illustrated in the inset. The spectra are shifted vertically for clarity. The symbols plot the respective decay times as a function of detection energy against the right axis.

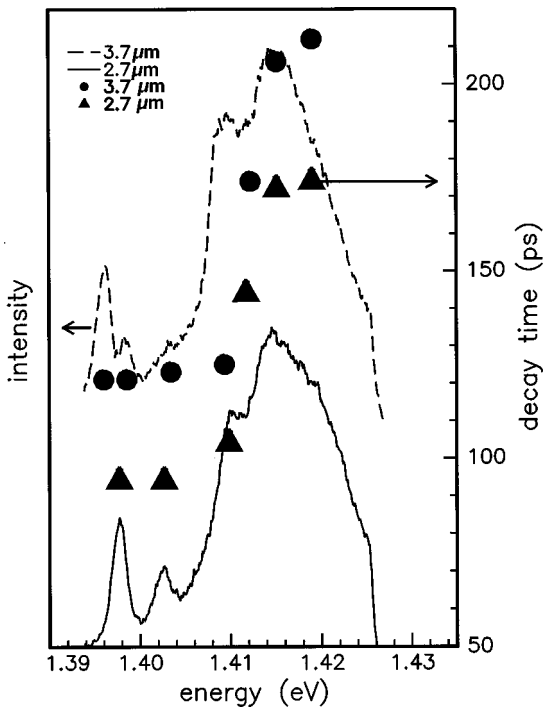

FIG. 2. Time-integrated spectra of 3.7 μm (dashed line) and 2.7 μm microcavities (full line). The symbols plot the respective decay times as a function of detection energy against the right axis.

the full cavity structures and create approximately the same carrier density in the $In_xGa_{1-x}As$ QW's.[14] The spectrum of the cavityless structure includes only the excitonic ground-state emission at $E_{ex}=1.415$ eV which is inhomogeneously broadened due to small variations in In content and width of the three QW's. In the case of the complete microcavities, full three-dimensional confinement is introduced on the electromagnetic field. The sharp emission lines E_i which additionally appear below the exciton line can thus be attributed to fully quantized photon modes with increasing lateral wave vectors, which are coupled to the excitonic transition. Their energetic positions systematically depend on structure size, as published elsewhere.[12]

The photoluminescence decay times of the two samples are given as a function of detection energy in Fig. 1 as well. The spontaneous emission of the exciton band follows a single-exponential decay which is considerably shortened from 309 ps (dots) to 173 ps (triangles) by placing the excitons inside the Bragg resonator. While the excitonic decay time is approximately constant over the whole exciton line of the cavityless sample it strongly decreases on the low-energy side towards the photon modes of the full microcavity structures. The photon modes are all characterized by similar and still faster decay constants of around 95 ps. This indicates that the interaction with the photon modes is responsible for the reduction of the overall excitonic decay time inside the microresonator.

Coupling between the exciton and photon modes should strongly depend on the detuning $\Delta_i=E_i-E_{ex}$ of the respective eigenenergies.[15] We have therefore investigated the photoluminescence kinetics for different structure sizes allowing us to vary the photon mode energies and hence the detunings. Figure 2 depicts the time-integrated spectra of 3.7 μm (dashed line) and 2.7 μm microcavities (full line). It is clearly seen that the photon energies are blue shifted and the energy splittings are increased for smaller structure size. Figure 2 also includes decay times determined for different detection energies on both microcavities (dots: 3.7 μm, triangles: 2.7 μm). The relative energy dependence is very similar in both microcavities. However, the absolute magnitude of the decay times is significantly smaller in the case of the smaller microcavities.

Figure 3 displays the measured decay time data as a function of structure size for both complete microcavities (open symbols and crosses) and structures without the upper Bragg mirror (filled squares), and compares them with the corresponding time constants of a quasi-two-dimensional mesa structure (lateral dimension of 100 μm). We observe a strong decrease of the exciton lifetime (diamonds) by a factor of nearly 6 when the lateral size of the complete microresonators is reduced from 100 μm (decay time of 560 ps) down to 1.7 μm (decay time of 100 ps). The decay time of the 2D mesa structure is approximately equal to the decay time of the QW emission for the mesa structure with a removed upper Bragg mirror. In contrast to the cavities with three-dimensional optical confinement, this implies that in the two-dimensional system there is no significant influence of the cavity on the overall exciton lifetime.

The exciton lifetime for structures with a removed upper Bragg mirror (filled squares) decreases only by a factor of 2 when the lateral size is reduced from 100 to 1.7 μm. Hence, the existence of a three-dimensional cavity strongly reduces

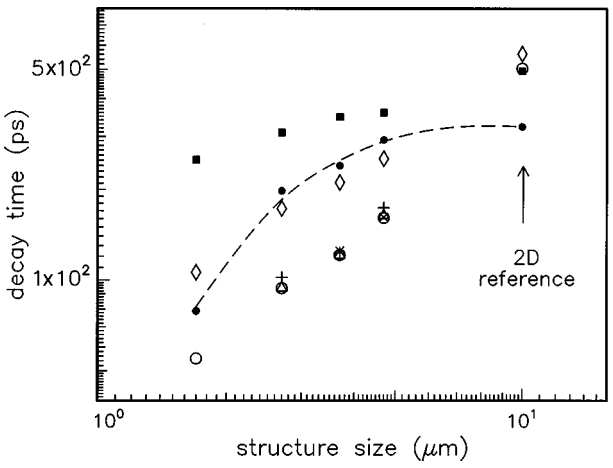

FIG. 3. Experimental decay times as a function of structure size for the cavityless structures (filled squares) and the complete microresonators (◇ : exciton, ○ : 1st, Δ : 2nd, + : 3rd, × : 4th cavity eigenmode). Calculated exciton lifetimes are given as dots (dashed line to guide the eye).

the exciton lifetime by up to a factor of 2.5 in the case of the smallest structure. Besides, nonradiative recombination does not significantly influence the lifetimes in the present rather wide structures with a full Bragg resonator (change of τ_{ex} by around 15%).

As shown in Fig. 3 the decay time of the photon mode of the mesa structure is only slightly smaller than its exciton lifetime. In contrast, the decay times of the photon modes in the three-dimensional cavities, which—remarkably—are all equal for a given structure size, are smaller than the corresponding exciton lifetime by approximately a factor of 2. i.e., the size dependence of the decay time measured at the energies of the photon modes is similar to the size dependence of the exciton lifetimes in the microresonators with three-dimensional optical confinement.

We first interpret the exciton decay time data. Coupling between the long-lived exciton (decoupled lifetime of $\tau_{ex,0}=550$ ps) and the short-lived photon modes intermixes the corresponding wave functions with the same in-plane wave vector. The exciton lifetime is decreased which we attempt to describe by a simple coupled harmonic oscillator model.[16] The radiative linewidth of the longer-lifetime (excitonlike) polariton mode takes, as a function of detuning Δ, the form

$$\gamma_{ex}=\hbar(2\tau_{ex})^{-1}=\tfrac{1}{2}[\Gamma-|\mathrm{Im}\sqrt{(\Delta+i\gamma)^2+\Omega^2}|], \qquad (1)$$

where $\Gamma=\gamma_{ph}+\gamma_{ex}$, $\gamma=\gamma_{ph}-\gamma_{ex}$, $\gamma_{ph}=\hbar(2\tau_{ph})^{-1}$, and $\gamma_{ex}=\hbar(2\tau_{ex})^{-1}$ are the homogeneous (radiative) half widths at half maximum of the decoupled photon mode and exciton, respectively. Ω is the Rabi splitting between the exciton and the examined photon mode. It has been measured for the different discrete photon modes and different structure sizes on a similar sample.[17]

In Eq. (1) we successively use the experimental detunings and the photon linewidths of the different photon modes *observed* in cw spectra with higher spectral resolution. Averaging over the obtained exciton lifetimes we can reproduce the decrease of the exciton lifetime for the 0D microcavities (dots in Fig. 3, dashed line to guide the eye). Careful investigation of Eq. (1) reveals that the decrease of the exciton lifetime is due to both, a smaller detuning between exciton and photon modes *and* smaller photon lifetimes for smaller structure size and higher photon modes. The latter are attributed to enhanced absorption at energetic positions closer to the exciton line and increased diffraction and scattering losses for smaller cavity sizes and higher modes.[18] For the same reasons higher energy modes are not observed. They couple only weakly to the exciton and do not influence the averaged exciton lifetime.

Note that for the 2D reference structure the calculated and measured exciton lifetimes do not agree at all. The existence of the continuum of leaky and guided modes prevents an influence of the planar cavity on the overall exciton lifetime.[5,6] In the three-dimensionally confined microresonator structures, however, the leaky and guided modes are expected to be considerably suppressed[19] which may therefore allow for a good description by a model including coupling to the resonant photon modes only.

In order to understand the decay times of the photon modes it has to be explained how part of the excitation energy can be transferred to the photon modes as far as 20 meV below the exciton emission line. First, we consider the coherent coupling between the photon and exciton modes which causes cavity polariton formation. Fast Rabi oscillations intermix the photon and exciton wave functions even in the weak coupling regime and allow the system to partly decay via the photon modes. In contrast to the long exciton lifetime the photon lifetime is hardly changed by the coherent coupling to the exciton. It is expected to be of the order of a picosecond as can be estimated from the photon linewidths of single microresonator structures.

The experimental decay times of the photon modes, however, are seen to be about two orders of magnitude larger than the expected photon lifetimes. In addition, the photon emission decay times closely follow the decrease of the exciton decay times with decreasing structure size. We therefore conclude that incoherent scattering processes between the exciton and photon modes play the dominant role in the photon modes dynamics: Due to the small excitonic admixture into the photon wave function the photon modes can be populated from the exciton branch via acoustic phonon or Auger-like scattering processes. The photon emission decay time will then be controlled by the longer exciton decay time through these relaxation processes.

We consider the Auger-like scattering process as a special exciton-exciton scattering process: One exciton is scattered into a photonlike state at lower energy while another exciton is simultaneously excited into an exciton state with higher energy. The exciton density excited per pulse is estimated to be 5×10^{10} cm^{-2} in our experiment, well below the critical Mott density. However, this density should allow for considerable exciton-exciton scattering as seen in four-wave-mixing experiments on a similar quantum well structure (approximately one exciton-exciton scattering process per picosecond).

If the Auger-type processes dominate the scattering into the photon modes the photon emission decay time is expected to be $\tau_{ex}/2$ due to a scattering rate which depends quadratically on exciton density.[20] As long as *all* photon

modes are predominantly populated via Auger relaxation processes this should hold for *all* photon modes of a given structure size independently of the different coherent coupling strengths of the photon modes with the exciton. It is precisely this behavior which is observed for the photon modes in Fig. 3. Further evidence for our interpretation is given by the time-integrated spectra: The intensity of the exciton increases linearly with the excitation power, the intensity of the photon modes with the square of the excitation power, which is anticipated in the case of a dominant quadratic relaxation term. It is this Auger-type relaxation process which increasingly populates the photon modes and eventually allows for stimulated emission.

Photon emission decay times slightly larger than $\tau_{ex}/2$ which are observed for the larger zero-dimensional structures are attributed to some contribution of phonon relaxation processes with transition rates depending linearly on exciton density. These processes are expected to determine the photon decay at longer delay times when the exciton density has considerably dropped and Auger-like relaxation has become weak. In fact, the time evolution of the photon modes is observed to decay with a time constant similar to τ_{ex} at late times.

Note finally that the photon emission decay time of the mesa structure is only slightly smaller than its exciton lifetime. In this context, we add that the strength of the linear acoustic phonon relaxation processes possibly depends on the cavity dimensions: In the two-dimensional cavity the polariton dispersion relation is a continuous function of the in-plane K vector. Therefore, carriers can easily relax down into the lower photonic branch via subsequent emission of low-energetic acoustic phonons. The discrete wave vectors in the zero-dimensional cavities lead to discrete photon modes which are separated by a few meV in energy. Acoustic phonon emission processes might therefore be considerably suppressed.

In conclusion, we have observed a strong enhancement of spontaneous emission rates by lateral photon confinement in Bragg microcavities. The exciton decay times decrease with lateral structure size mainly due to coupling with the short-lived photon modes. The decay times of the photon mode emission are shown to reflect neither the photon lifetimes, nor the different coherent coupling strengths with the exciton. Incoherent Auger-type scattering processes can account for photon emission decay times of approximately half of the corresponding exciton lifetime for *all* photon modes of a given 0D microstructure.

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