

Cavity-Induced Changes of Spontaneous Emission Lifetime in One-Dimensional Semiconductor Microcavities

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Spontaneous emission lifetime in a one-dimensional AlGaAs distributed Bragg reflector microcavity has been systematically measured as a function of emission wavelength tuned by the quantum-confined Stark effect in order to confirm the cavity-induced alteration of the SE lifetime. The result manifests a substantial cavity-induced fractional change of the exciton decay times, $\sim 30\%$ around the on-axis resonant wavelength, despite the relatively large broadening of the excitonic emission being comparable to the cavity resonance width in the low- Q cavity.

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Much attention has been paid to the observation of cavity-induced changes of spontaneous emission (SE) electron lifetime in order to confirm the existence of the cavity effect, because the lifetime changes are the most essential effect in the original concept of the modification of spontaneous emission proposed by Purcell [1]. Although there have been several experiments demonstrating the modifications of the SE carrier lifetime in semiconductor microcavities [2–6], there exist very few systematic studies and quantitative comparison with theory. This is because the rate of change of the SE lifetime in one dimensionally confined microcavity structures has been predicted theoretically to be relatively small [7–11].

The requirements for detection of such cavity-induced SE lifetime changes in one-dimensional cavities are as follows [5]. First, the optically active media must have very high internal quantum efficiencies. Second, the cavity quality factor Q has to be high enough to observe the cavity-induced SE lifetime changes but low enough for photon reabsorption to be negligible [5,12]. A system satisfying the above requirements has been realized recently using an Er-doped transparent Si/SiO₂ planar microcavity [5], which provided a quantitative comparison between the measured SE rates as a function of wavelength detuning and the theoretical result.

In this Letter, we report the measured cavity-induced SE lifetime changes in semiconductor microcavities using an alternative and simpler scheme in which the emission wavelength, instead of the changing resonant wavelength, is tuned by the quantum-confined Stark effect (QCSE) under a fixed cavity resonant characteristic. This method seems to be preferable for carrying out a systematic study on the cavity-induced SE lifetime changes specifically in semiconductor microcavities. In particular, the following point will be clarified by the demonstration of our experimental results. The fraction of the SE (the so-called coupling coefficient of SE, denoted as β_c in this Letter) coupled into *one* dominant planar cavity mode is, in general, small, i.e., of the order of 10^{-2} for

the emission broadening comparable to or larger than the cavity resonance width [13]. Consequently, at first sight, one may conclude that the fractional change in the SE rates should also be small unless a very large cavity Q value and very small emission broadening are realized. This is not true in the cases of planar cavities. The density of states of planar cavity modes feature a quasicontinuum for the emission wavelength shorter than the on-axis resonant wavelength. Hence the overall coupling of the SE into many quasicontinuum planar cavity modes distributed over the wavelength range of the emission spectrum may result in a substantial change in the SE rates. Thus, the fractional change in the SE rates is expected to be larger than β_c , independent of the cavity Q value and emission broadening in planar cavities unless the Q value and broadening are extremely low and large, respectively. In other words, the Q value and emission broadening are not essential for the total amount of fractional change in the SE rates although the spectral width of the fractional change may depend on the Q value and emission linewidth. This important point has not been widely recognized to date and will be demonstrated experimentally in this Letter, manifesting a large cavity-induced fractional change of the exciton decay times, $\sim 30\%$ in semiconductor planar cavities.

The vertical structure of the microcavity tested in the present experiment was grown by a molecular beam epitaxy technique on an n -doped GaAs substrate [6,14]. An undoped GaAs single quantum well (QW) with a thickness of 10 nm is placed at the center of an undoped AlAs optical cavity with a half-wavelength (~ 130 nm) thickness, which is sandwiched by two (top and bottom) distributed Bragg reflectors (DBR's) composed of p - and n -doped 10.5 alternative quarter-wavelength layer pairs of Al_{0.2}Ga_{0.8}As and AlAs, respectively. In such a sample, spontaneous emission is expected to be enhanced on resonance where vacuum field intensity is maximized at the QW layer.

The photoluminescence (PL) dc measurement was performed at 12–50 K by selectively as well as nonreso-

nantly pumping the GaAs single QW with a semiconductor laser (Hitachi HL7852G laser diode) with a photon energy of 1.58 eV, much lower than the band gap of $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ (~ 1.763 eV at 50 K) and slightly higher than the exciton gap of the QW, at low temperatures below 50 K. The pump power of ~ 13 mW used in the PL measurements, which was focused onto a spot size of $\sim 10^4 \mu\text{m}^2$, was expected to generate a carrier density of the order of $\sim 10^{15} \text{cm}^{-3}$ under the postulation of a carrier lifetime of ~ 1 ns. Such an excitation level guarantees that an emission from the sample is a spontaneous one, not a stimulated one. The postulated lifetime ~ 1 ns will be justified later by the result of time-resolved PL decay measurements.

The PL was collected along the cavity axis within a solid angle of $\sim 7 \times 10^{-2} \pi$ measured from the cavity axis. The redshift of the peak emission wavelength due to QCSE with increasing internal fields was clearly observed, and when the peak emission wavelength λ_e was tuned to the on-axis resonant wavelength $\lambda_r \sim 801\text{--}809$ nm the PL intensity was drastically enhanced compared with PL intensity per QW layer measured with a GaAs/AlAs multiple quantum well structure with no cavity, which is considered to be a free space emission [6,14,15].

A simple criterion for negligible photon reabsorption is given by $\alpha L_z \ll 1 - R$ where L_z is the thickness of an absorbing medium (QW in the present case), α is the absorption coefficient for the absorbing medium, and R is the cavity mirror reflection coefficient [16,17]. The present system with $R \sim 0.88$ and $\alpha \sim 10^4 \text{cm}$ for the 100 Å QW fulfills the above criterion so that photon reabsorption can be negligible. This was obvious by the negligibly weak dependence of measured reflection spectra on applied voltages [15].

PL decay measurement was performed for the sample, named sample A, with the resonance wavelength $\lambda_r \sim 809$ nm at 30 K as a function of the peak emission wavelength which was continuously tuned by the QCSE [6]. The sample was excited by 300 ps light pulses emitted from the same laser diode as in the PL dc measurement. In Fig. 1, open circles show the measured PL decay times $\tau(\lambda_e)$ as a function of the peak emission wavelength for optical transitions between the lowest quantized conduction band and the lowest quantized heavy hole band ($1e\text{-}1hh$) in the GaAs QW. An increase in the PL decay time was observed with the redshift of the peak emission wavelength, which is due to the cavity effect as well as QCSE. A sudden decrease in the PL decay time for $\lambda_e \sim 809$ nm is attributed to photoexcited carrier leakage from the GaAs QW by an intense electric field by the fact that the measured photocurrent began to increase around $\lambda_e \sim 809$ nm as shown in Fig. 1. In order to obtain a pure change of the PL decay time due to cavity effect, $\tau_c(\lambda_e)$, the QCSE-related change of the PL decay time in the region of $\lambda_e < 809$ nm was removed by multiplying $\tau(\lambda_e)$ with the

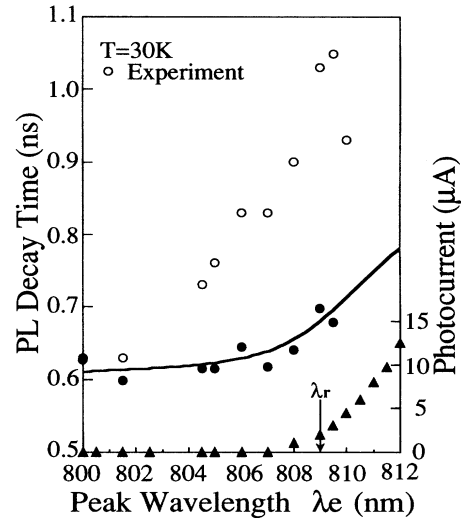


FIG. 1. ○: The measured PL decay times as a function of the peak emission wavelength for sample A with an on-axis resonant wavelength of $\lambda_r = 809$ nm at $T \sim 30$ K. ●: The cavity-induced PL decay time changes obtained by theoretical separation of the change of the lifetime due to QCSE from the data (○). Solid line: The calculated variation of the SE lifetime with the emission wavelength due to cavity effect. ▲: The measured photocurrent data.

calculated squared overlap integral between the lowest electron and the lowest heavy hole wave functions, $M_{1e-1hh}^2(\lambda_e)$ since $\tau(\lambda_e) = \tau_c(\lambda_e)/M_{1e-1hh}^2(\lambda_e)$ [18]. The validity of such a way of separating the change due to QCSE has been already confirmed in previous work [6]. The obtained τ_c 's are plotted and compared with the calculated variation of the SE lifetime with the emission wavelength in the planar DBR microcavity [8], showing fairly good quantitative agreement. In this calculation, all optical dipole moments are assumed to be along the QW plane [19] and excited electron-hole pairs are considered to form pointlike excitons [8] without coherent effect [20,21]. Moreover, the full width at half maximum (FWHM) of the spontaneous emission line, $\delta\lambda_e = 3$ nm, obtained by the PL measurement is included in all calculations shown in this Letter. Note the following points: (a) the overall coupling of the SE into many cavity modes discussed in the introductory part of this Letter is correctly counted by the simple inclusion of the emission broadening into the theoretical treatment developed in Ref. [8] and (b) it is not necessary to distinguish between the homogeneous and inhomogeneous broadenings in the present discussion. However, because of the presence of nonradiative processes due to carrier leakage around $\lambda_e = \lambda_r$, the cavity-induced changes for the region of $\lambda_e > \lambda_r$ were not obtainable.

Figure 2 shows the peak emission wavelength dependence of PL decay time, $\tau_c(\lambda_e)$, given by another sample named sample B which was obtained by cleaving a differ-

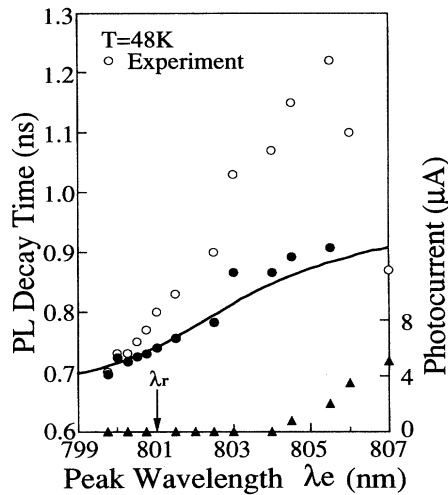


FIG. 2. The measured PL decay times as a function of the peak emission wavelength at $T \sim 48$ K for sample B with $\lambda_r \sim 801$ nm which is shorter than that of sample A. All notations are the same as in Fig. 1.

ent part of the same wafer as sample A. In general, due to the nonuniformity of growth rate, spatial fluctuations of the on-axis resonant wavelength over a wafer are more or less present [3]. In the present case, the on-axis wavelength of sample B is ~ 801 nm, which is shorter than that of sample A. As a result, the cavity-induced change of the PL decay time, $\tau_c(\lambda_e)$, for the region of $\lambda_e > \lambda_r$, which was not obtained with sample A, was obtained.

According to the theoretical prediction of the emission wavelength dependence of the SE lifetime in planar DBR microcavities, the lifetimes sharply increase with increasing emission wavelength around $\lambda_e \sim \lambda_r$ and gradually increase in the regions of $\lambda_e < \lambda_r$ and $\lambda_e > \lambda_r$. As shown so far, there is a difficulty in observing the whole structure of the cavity-induced SE lifetime changes with a single sample because the occurrence of nonradiative processes due to carrier leakage induced by intense electric fields limits the available tunable range of the emission wavelength. Thus we compile the data of PL decay times for four different samples including sample A and sample B as shown in Fig. 3 where the decay times normalized to the decay time at λ_r , $\tau_c(\lambda_e)/\tau_c(\lambda_r)$, are plotted versus the normalized wavelength λ_e/λ_r for each sample. The obtained τ_c 's manifest a steplike feature of the cavity-induced SE lifetime changes which is anticipated theoretically.

As shown in the table of Fig. 3, the absolute values of the decay times are sample dependent. One candidate for the possible reasons for this is the presence of nonradiative recombination processes. However, nonradiative recombination processes would not be responsible for the sample-dependent carrier lifetimes because high internal quantum efficiencies were confirmed for all samples both by the observed linear dependence between output power and pump power and by excellent agreement between the

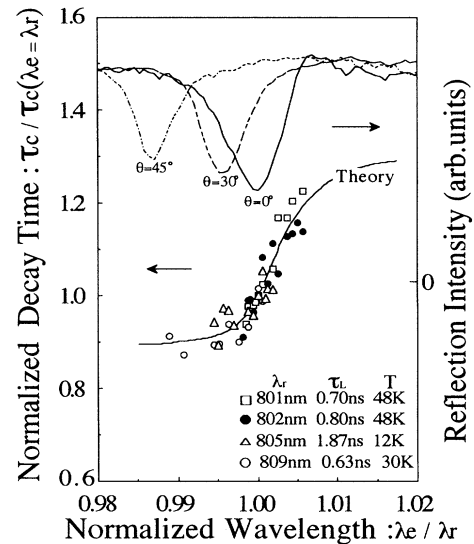


FIG. 3. The normalized cavity-induced PL decay time $\tau_c(\lambda_e)/\tau_c(\lambda_e = \lambda_r)$ vs the normalized wavelength λ_e/λ_r for four different samples with different on-axis resonant wavelengths λ_r . The value of λ_r , temperature of the measurement T , and the measured shortest decay time τ_L for each sample are shown. Reflection spectra for various angles of $\theta = 0^\circ$, 30° , and 45° measured from the cavity axis are also shown.

measured transfer efficiency and the theoretical value obtained by assuming that nonradiative processes are absent [15]. Another candidate is the coherent exciton effect by which the radiative recombination rate is enhanced by a factor of $A_c/\pi a_{2D}^2$ where a_{2D} is the Bohr radius of a (quasi)-two-dimensional exciton and A_c is the area of exciton coherency [20,21]. According to Feldmann *et al.* [20], the coherent area A_c for localized excitons is determined by the extent of the localized excitons and that for free excitons is determined by the exciton scatterings due to phonon, impurity, and interface scattering. Judging from the fact that the measured shortest decay time τ_L obtained with zero electric field, shown in the table of Fig. 3, did not show a systematic temperature dependence, the observed sample-dependent decay times could be attributed to the coherent area fluctuations mainly caused by interface scattering for free excitons and island area fluctuations for localized excitons, which both result from the well-width fluctuation, i.e., by inhomogeneous contributions rather than homogeneous contributions due to phonon scattering. It is important to point out here that, by inference from the absolute values of the measured decay times, the decay rate enhancement due to the coherent exciton effect may not be so significant, indicating that $A_c \sim \pi a_{2D}^2$. This explains the fact that the observed change of the decay time with the emission wavelength is in excellent agreement with the theoretical result in which pointlike excitons are assumed [8].

Let us examine such a steplike dependence of the SE lifetime on the emission wavelength in more detail. In

the region of $\lambda_e < \lambda_r$, a lifetime reduction occurs. This is because for $\lambda_e < \lambda_r$ the resonant condition is always satisfied at an internal offset angle $\theta_{in} = \cos^{-1}(\lambda_e/\lambda_r)$ measured from the cavity axis, i.e., the corresponding external emission angle $\theta_{ex} = \sin^{-1}(n_{eff} \sin \theta_{in})$ where n_{eff} is the effective refractive index of a microcavity. The existence of the off-axis resonant modes is confirmed by the measured surface reflection spectra for various oblique incident angles shown in Fig. 3. As a result, the total coupling of SE into many quasicontinuum off-axis modes leads to a substantial reduction of the exciton decay times. On the other hand, when $\lambda_e > \lambda_r$, lifetime enhancement occurs because the resonance condition is not satisfied by any angles.

A detailed theoretical study reveals that the fraction $\beta_t \sim 6\%$ of the total emission rate contributes to the main lobes on the air side and 24% contributes to those on the substrate side, where “main lobe” means emissions within a small solid angle around a cavity axis, caused by main planar cavity modes. The asymmetry of β_t is due to the reflectivity difference between the air and substrate side DBR’s, a difference which is caused by a large discontinuity of the refractive index at the interface between air and AlGaAs. The fraction of the total emission rate named the transfer efficiency of the SE, β_t , should be strictly distinguished from the coupling efficiency of the SE, β_c . β_t includes the excited quasicontinuum of modes to the blue side of the on-axis resonance in addition to the dominant mode which would lase first. All the excited quasicontinuum modes bring the emissions within the small solid angle of the “main” lobe. On the other hand, the fraction of SE’s emitted into the “lasing” mode determines β_c . For the enhancement of β_t , it is not strictly required for the SE linewidth to be smaller than the cavity linewidth. This is because the emitted photons which belong to any main cavity modes can contribute to the transfer efficiency, no matter which specific modes they belong to. Thus the transfer efficiency β_t (not coupling efficiency β_c) should be correlated to the fractional change of the SE rates. The estimated transfer efficiency, 6% on the air side, is consistent with the measured external quantum efficiency for the main lobe emission on the air side, $\sim 5\%$ [15]. Therefore, the observed 30% increase in the lifetime around $\lambda_e \sim \lambda_r$ could be ascribed to suppression of the main lobe emissions on both air (6%) and substrate (24%) sides.

In conclusion, we have systematically studied the cavity-induced spontaneous emission lifetime changes in a one-dimensional AlGaAs distributed Bragg reflector microcavity by virtue of the continuous tuning of the emission wavelength by the quantum confined Stark effect. It should be noted that the large fractional change in the lifetime has been obtained with the relatively large broadening of the excitonic emission, ~ 3 nm, comparable to the cavity resonance width of low- Q cavities.

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