Inhibition and Enhancement of the Spontaneous Emission of Quantum Dots in Structured Microresonators

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Spontaneous emission of quantum dot systems in laterally structured microcavities that exhibit photon confinement in all three directions has been studied by time-resolved photoluminescence spectroscopy. For on-resonance conditions, we find that the dot emission rate is increased substantially over that of the unstructured planar cavity. For off-resonance conditions, we are able to suppress the emission rate by an order of magnitude by using cavities with metal coatings, which we attribute to the suppression of leaky optical modes in these structures.

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Since its proposal by Purcell [1] and early work involving atoms in cavities [2], "cavity quantum electrodynamics" (CQED) has been actively pursued for its potential insight into fundamental problems in light matter interaction. Here the energies and modes of photon states, to which the electronic transitions of atoms can couple, are changed by the cavity confinement. Recently, efforts have been made to extend this work to the optical regime, which requires cavities with sizes on the order of microns. This work has been made possible by the epitaxial fabrication of high quality layered semiconductors [3] from which optical cavities have been made that have strong photonic confinement in one direction. Quasiatomic light emitters in these resonators can be mimicked by semiconductor quantum dots (QDs) that exhibit a discrete density of electronic states.

A central aspect of cavity research over the years has been the possibility to modify spontaneous emission rates [1,4] in the regime of weak coupling between the electronic and photonic excitations which opens opportunities for basic physics studies as well as in semiconductor optoelectronics. For "on resonance" conditions, where the electronic transition energy coincides with the cavity mode energy, the emission rate can be increased. On the other hand when the electronic transitions are "off resonance," the emission can be suppressed. These features are of interest, e.g., in connection with bright light emitting diodes and low threshold lasers or in implementations of quantum computing or quantum cryptography: In the case of quantum cryptography, for example, single photon devices require a detailed control of spontaneous emission.

In planar cavities only modest enhancements and suppressions of emission rates on the order of 10% have been observed [5,6]. Very recently, for laterally structured cavities that exhibit photon confinement in all three directions enhancements in the on-resonance emission by a factor of up to 5 have been demonstrated [7]. However, in these structures it was not possible to demonstrate a suppression of the off-resonance emission. This was attributed to the confined optical modes being superimposed on a quasicontinuum of "leaky" photon modes along the cavity [8]. As a result, the off-resonance emission is little changed from its value in a homogeneous medium.

In the present work we have developed a new type of cavity in which the lateral sidewalls of the resonator are coated with metal to suppress the off-resonance leaky modes. We show that these cavities enhance the on-resonance emission in a manner similar to the uncoated laterally structured cavities. In addition, under off-resonance conditions we find a strong increase of the emission lifetime, which for the first time gives evidence of a significant inhibition of the spontaneous emission in a solid state system.

The resonators used in this work are based on a standard wedge-shaped λ cavity consisting of a GaAs resonator sandwiched between high reflectance GaAs/AlAs Bragg mirrors. The finesse $Q = \lambda_C/\Delta\lambda_C$ of the planar resonators is $\sim 7.0 \times 10^3$. From these cavities cylindrical microresonators with diameters ranging from 3 to 5 μm have been fabricated by lithography and etching [9,10]. For one type of structure the lateral sidewalls of the resonators were left uncovered after the patterning. For a second type of structure the sidewalls were coated by a gold layer with a thickness of $\sim$150 nm.

QD emitters form the optically active medium in these cavities. These quasi-zero-dimensional systems are obtained by placing a strongly inhomogeneously broadened $In_{0.14}Ga_{0.86}As$ quantum well with a nominal width of 7 nm in the center of the cavity. The lower trace in Fig. 1(a) shows a photoluminescence spectrum of this quantum well recorded on a sample from which the top mirror was removed. The diameter of the exciting laser spot was $\sim$20 μm. The full width of the inhomogeneously broadened emission line is about 15 meV. By exciting with a microscope objective, the laser spot size could be reduced to $\sim$1.5 μm. The upper trace in Fig. 1(a) shows the luminescence spectrum recorded under such excitation conditions. The broad emission band is broken into a set of sharp

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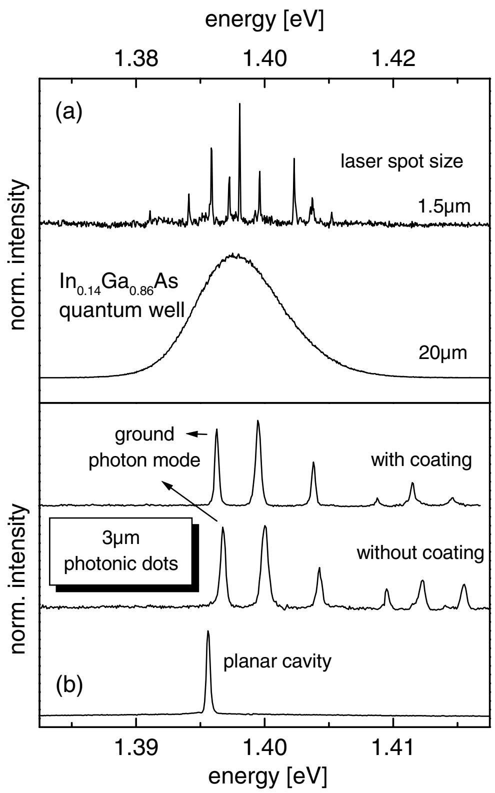

FIG. 1. (a) Photoluminescence spectra of a strongly inhomogeneously broadened $In_{0.14}Ga_{0.86}As/GaAs$ quantum well. The lower trace shows a spectrum for which the diameter of the exciting Ar^+ laser spot was ~ 20 μm. The upper trace is a spectrum recorded with a spot diameter of about 1.5 μm. (b) Luminescence spectra of 3 μm sized photonic dots with uncoated and coated lateral sidewalls in comparison to the spectrum of a planar resonator.

emission lines with widths <50 μeV. Thus the quantum well structure can be considered as an ensemble of potential minima that are separated from each other by lateral barriers [11,12]. The sharp emission lines originate from the recombination of electron-hole pairs trapped in these QD minima.

Figure 1(b) shows luminescence spectra of patterned resonators with diameters of 3 μm compared to the spectrum of a planar cavity. For the planar structure the spectrum is dominated by the QD emission into the cavity mode on a weak background arising from the dot emission into leaky modes. For both the coated and uncoated resonators, the QD emission into several confined optical modes is observed [9,10]. The energy separations between them are equal for the two structures. For the uncoated structures we showed recently that the electromagnetic fields are well localized in the resonators for structure sizes >1 μm due to the large discontinuity of the refractive index between semiconductor and vacuum [13]. This dielectric discontinuity results in a sidewall reflectivity of ~ 0.35 for normal incidence. For the metal coated structures the reflectivity there tends to unity. Corresponding calculations for these coated cavities show that the modes are also strongly confined in them and that the energies are essentially the same in the two types of cavities.

The kinetics of the QD emission from the resonators has been studied by time-resolved photoluminescence spectroscopy at $T = 2$ K on arrays of uncoated and coated resonators. For quantitative comparison, resonators of comparable finesses were investigated for both structure types. The maximum achievable Q factors were 6.9×10^3 for the 5 μm, 6.7×10^3 for the 4 μm, and 6.4×10^3 for the 3 μm sized cavities. Optical excitation was done by a mode-locked Ti:sapphire laser emitting pulses with a length of 1.5 ps at a repetition rate of 82 MHz. The laser energy was tuned to 1.467 eV to excite the InGaAs only. Time-averaged excitation powers of less than 2 mW were used to avoid nonlinear optical effects. The radiation emitted within an angle of 60° relative to the cavity normal was collected, spectrally analyzed by a monochromator ($f = 0.32$ m) and detected by a streak camera. The temporal resolution of the setup was about 10 ps.

Figure 2 shows photoluminescence decay curves for uncoated (upper panel) and coated (lower panel) resonators with a diameter of 3 μm for different detunings from the ground photon mode. The decay is taken at an energy E

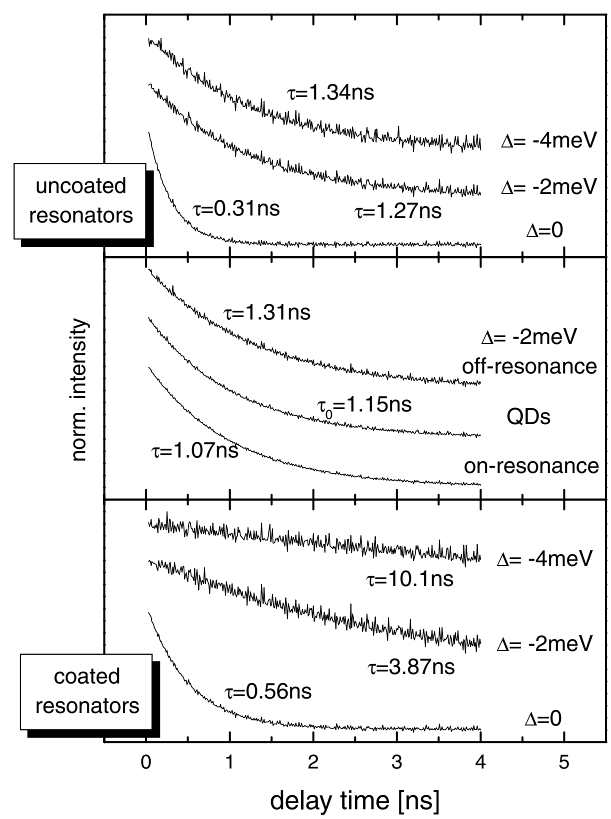

FIG. 2. Photoluminescence decay curves of uncoated (upper panel) and coated (lower panel) resonators with a diameter of 3 μm for various detunings Δ from the ground optical mode (indicated at each trace). The middle panel shows the decay curves for the QDs with the upper mirror removed (middle trace) and also for the planar cavity for on and for off resonance.

that is separated by $\Delta = E - E_C$ from the lowest cavity mode at E_C. The decay curves for the QDs with the top mirror removed from the cavity and for planar resonators also are shown (middle panel). For short delays (<50 ps) scattered laser light appears in the spectra, so only the data for later times are shown. In all cases the luminescence decays to a good approximation exponentially. For the QDs without cavity the luminescence lifetime is ~1 ns. In agreement with previous reports [5,6], in the planar resonator only marginal changes of this lifetime are observed, for both on and off resonance.

For off resonance in the uncoated resonators, the lifetimes are as well only slightly increased over the QD value. However, for on resonance the lifetime is considerably shortened to $\tau \sim 0.3$ ns [14]. A similar shortening to $\tau \sim 0.5$ ns is observed for the coated structures [14]. But, in contrast, for these structures the off-resonance lifetime increases drastically: For $\Delta = -2$ meV, $\tau \sim$ 3.9 ns, while for $\Delta = -4$ meV, $\tau > 10$ ns.

Figure 3 shows the photoluminescence decay rates τ_0/τ as functions of detuning Δ from the ground photonic mode for the 3 μm resonators. Here the decay rate $1/\tau$ is normalized by that of the QDs without cavity $1/\tau_0$. For comparison, this ratio for the planar cavity is also plotted there. In this case, the data show that the QD emission dynamics can be altered by only about 10% by the resonator for both on and off resonance, in agreement with previous reports [5,6]. This also holds for the off-resonance situation in the uncoated patterned resonators. For them, the emission rate still is about 0.9 of that of the QD emission, independent of the detuning. On the other hand, for on resonance it can be enhanced by a factor of 3. A somewhat weaker enhancement by a factor of 2 is observed for the coated cavities. For off resonance, however, a considerable reduction of the emission rate is obtained here. For example, for $\Delta = -2$ meV, $\tau_0/\tau = 0.25$ and for $\Delta < -3$ meV, $\tau_0/\tau \sim 0.1$.

Figure 4(a) shows the decay rates τ_0/τ vs resonator size for on resonance of excitons and photons in the uncoated and coated resonators. For both types, an increase in the rate with decreasing resonator size is observed, which is larger for the uncoated resonators than for the coated ones. The size dependences can be described well by the form $c_1 + c_2/L^2$ where L is the lateral resonator size. The normalized decay rates τ_0/τ for being off resonance are plotted vs resonator size in Fig. 4(b). For the coated structures, the data for $\Delta = -2$ (open squares) and -4 meV (full squares) are shown, for the uncoated structures only

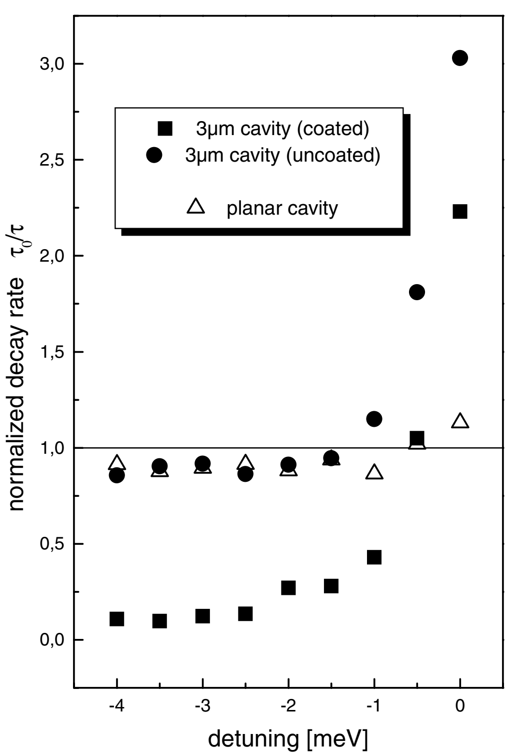

FIG. 3. Ratio of the spontaneous emission rates τ_0/τ of QDs in a cavity to that in a homogeneous medium versus detuning from the ground photon mode towards lower energies. The data for 3 μm resonators are compared with the data of a planar cavity.

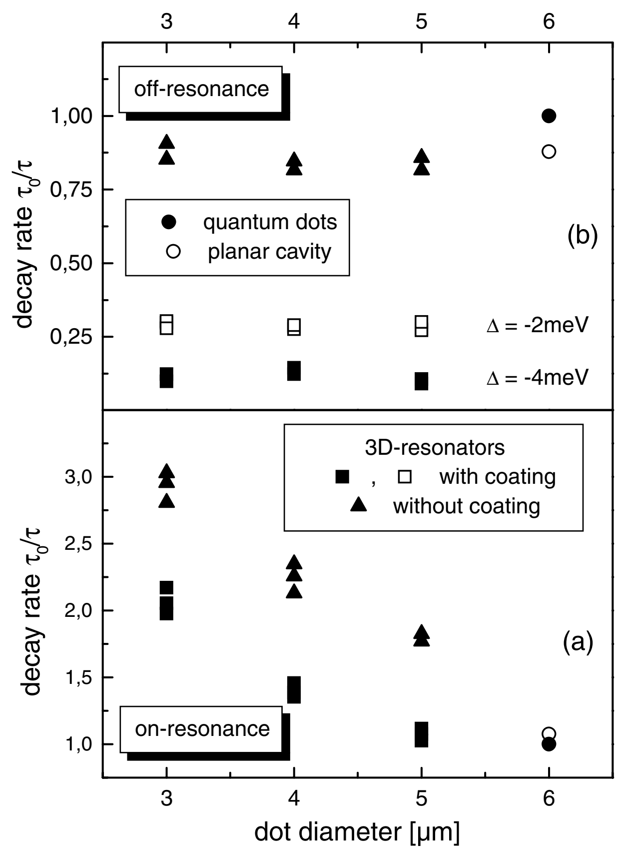

FIG. 4. Ratio of the spontaneous emission rates τ_0/τ of QDs in a cavity to that in a homogeneous medium (no cavity) versus lateral resonator size. (a) shows the data for on resonance, (b) LU7776for off resonance of excitons and photons. Also the decay rates for the QDs without cavity and for the QDs in a planar resonator are shown. For off resonance the decay rates at two different detunings Δ from the optical mode are plotted.

the data for $\Delta = -4$ meV. For both resonator types the observed decay rates show no dependence on resonator size. The most important result is the suppression of the emission rate by a factor of 10 in the coated cavities for large detunings ($\Delta = -4$ meV).

The emission dynamics of the QDs in the resonators is described in the weak coupling regime [7]: The linewidth $\Delta\lambda_E$ of the light emitting QDs is considerably smaller than the width of the cavity mode $\Delta\lambda_C$. Thus the emitter effectively couples to an optical mode continuum, and photon reabsorption can be neglected. In this dissipative regime the emission kinetics is given by Fermi's golden rule: The spontaneous emission rate $1/\tau$ of an emitter located at position $\mathbf{r}$ in the resonator relative to a location in a homogeneous medium $1/\tau_0$ is given by [7]

$$\frac{\tau_0}{\tau} = \frac{2}{3} F_P \frac{|\varepsilon(\mathbf{r})|^2}{|\varepsilon_{\max}|^2} \frac{\Delta\lambda_C^2}{\Delta\lambda_C^2 + 4(\lambda_C - \lambda_E)^2} + \alpha . \quad (1)$$

Equation (1) describes two different decay channels: The first term gives the QD emission into the cavity mode that has an electric field distribution $\varepsilon(\mathbf{r})$ with a maximum amplitude $\varepsilon_{\max}$. The Purcell factor F_P is given by

$$F_P = \frac{3}{4\pi^2} Q \frac{\lambda_C^3}{n^3 V} . \quad (2)$$

Here n is the refractive index of the resonator, and V is the effective volume of the optical mode. The second term α describes the decay channel due to QD emission into leaky modes.

For a given size the confinement of the fields of the lowest electromagnetic modes is essentially identical in the coated and uncoated resonators. Because we study cavities of comparable finesses, we expect the same dependences of the first decay channel in Eq. (1) for them. Then the difference between the decay rates of the uncoated (U) and coated (C) cavities, $\Delta\alpha = \alpha_U - \alpha_C$, is given by their different couplings to the leaky modes, for both on and off resonance. From the data in Fig. 3 we find $\Delta\alpha = 0.73 \pm 0.07$, and this difference does not depend on detuning within the experimental accuracy.

For off resonance the first decay channel in Eq. (1) can be neglected in comparison to the second one in the uncoated structures and the emission dynamics is predominantly given by the coupling to the leaky photon modes. It is the coupling to these modes which prevents a strong suppression of the QD spontaneous emission in these resonators. For the coated structures, in contrast, a suppression of this decay channel by an order of magnitude is found for $\Delta < -2$ meV. We note that based on the detuning dependence of the first term in Eq. (1) an even stronger deenhancement than the observed one might be expected for large detunings. From this we conclude that $\alpha_C \neq 0$, and the coatings do not fully suppress the leaky modes. In addition, absorption in the gold coatings might play some role. Further technological improvement of the coatings might allow for an even greater suppression.

For on resonance in the coated structures the QD emission is dominantly directed into the ground optical mode. For the uncoated structures the first decay channel becomes more and more efficient with decreasing structure size. The increase of the emission rates with decreasing L results from the Purcell effect, and is given by the reduction of the mode volume in F_P. Since the reduction of the cavity finesse is weak and can be neglected in first approximation, the emission rate is controlled by the confinement of the electrical field which goes approximately as $1/L^2$, in good agreement with the experimental data.

In summary, we have demonstrated both of the two critical aspects of the control of spontaneous emission in laterally structured semiconductor microcavities, a strong suppression of the off-resonance emission as well as a strong enhancement of on-resonance emission. To do this, we have introduced a new type of cavity with metal coated sidewalls which suppresses coupling to leaky modes off resonance. This progress will open new opportunities in studies of the optical properties of solids in which basic CQED effects in the optical regime can be explored in new detail down to the single photon level. It could also have impact in the suppression of decoherence due to spontaneous emission in implementations for quantum computation based on excitons in semiconductor microcavities with QDs [15].

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