Enhanced Spontaneous Emission by Quantum Boxes in a Monolithic Optical Microcavity

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(Received 3 April 1998)

Semiconductor quantum boxes (QB's) are well suited to cavity quantum electrodynamic experiments in the solid state because of their sharp emission. We study by time-resolved photoluminescence InAs QB's placed in the core of small-volume and high-finesse GaAs/AlAs pillar microresonators. A spontaneous emission rate enhancement by a factor of up to 5 is selectively observed for the QB's which are on resonance with one-cavity mode. We explain its magnitude by considering the Purcell figure of merit of the micropillars and the effect of the random spatial and spectral distributions of the QB's. [S0031-9007(98)06704-0]

PACS numbers: 78.55.Cr, 12.20.–m, 78.66.–w

In a founding paper, Purcell proposed to control the spontaneous emission (SE) rate of a quasimonochromatic dipole by using a cavity to tailor the number of electromagnetic modes to which it is coupled [1]. Cavity quantum electrodynamics (CQED) has provided a firm theoretical and experimental basis for this revolutionary concept [2] and has been since 1990 a major source of inspiration for the research activity on solid-state optical microcavities [3–14]. An ability to enhance the SE rate (Purcell effect) would open novel avenues for physics and engineering [3], e.g., the development of high-efficiency light emitters.

Let us first examine the requirements for observing a large Purcell effect. Originally [1], Purcell considered a localized dipole (wavelength λ_e, linewidth $\Delta\lambda_e$) placed on resonance with a single cavity mode (wavelength λ_c, linewidth $\Delta\lambda_c$, quality factor $Q = \lambda_c/\Delta\lambda_c$). Since $\Delta\lambda_e \ll \Delta\lambda_c$, the escape time of SE photons out of the cavity is much shorter than the radiative lifetime and reabsorption is negligible. In this so called "weak coupling regime," the emitter feels a quasicontinuum of modes and the SE rate is given by the Fermi golden rule. For an electric dipole transition

$$\frac{1}{\tau} = \frac{4\pi}{\hbar}\,\rho_{\rm cav}(\omega)\langle|\vec{d}\cdot\vec{\varepsilon}(\vec{r})|^2\rangle,$$

where $\vec{\varepsilon}(\vec{r})$ is the vacuum electric-field amplitude at the location $\vec{r}$ of the emitter and $\vec{d}$ is the electric dipole. $\rho_{\rm cav}(\omega)$ is the density of modes at the emitter's angular frequency ω and is given by a normalized Lorentzian in the cavity case. A simple derivation shows that the SE rate in the cavity mode, referenced to the total SE rate in a homogeneous medium, is given by the Purcell factor $F_p = 3Q\lambda_c^3/4\pi^2 n^3 V$, where n is the refractive index of the medium and V is the effective mode volume [15], provided several important conditions are satisfied: The emitter must be on exact resonance, located at the antinode of the vacuum field, with its dipole parallel to the vacuum electric field. In other words, the Purcell factor is a figure of merit for the cavity alone, which describes its ability to increase the coupling of an ideal emitter with the vacuum field, via a local enhancement of its intensity (small V's) or of the effective mode density (high Q's). In practice, however, the choice of the emitter is essential, since its linewidth puts an upper limit on the cavity Q's which can be used ($Q < \lambda_e/\Delta\lambda_e$). Early CQED experiments have benefited in this respect from the compatibility of Rydberg atoms and high-F_p microwave cavities, resulting in a 500-fold cavity enhancement of the SE rate [2].

At optical frequencies, planar dielectric cavities using distributed Bragg reflectors (DBR's) have been studied first due to the excellent control provided by layer-by-layer fabrication techniques. However, such planar cavities support a continuum of modes and theory [4,6] predicts SE enhancement factors close to unity for independent light emitters in the weak coupling regime, as observed, e.g., for rare-earth atoms [5] or GaAs quantum wells (QW's) in low Q cavities [6]. More recently, progress in microfabrication techniques has allowed a three-dimensional (3D) engineering of the refractive index on the wavelength scale and a rich diversification of the microcavity designs. Silica microspheres [8], pillar microresonators [9–11], photonic disks [12] and wires [13], and 1D photonic bandgap microcavities [14] sustain a discrete set of resonant modes (as well as a continuum of leaky modes) and have the potential to display the Purcell effect ($F_p \gg 1$), provided a convenient emitter is used. The large spectral width of commonly used emitters, i.e., rare-earth atoms in dielectric matrices (~20 nm at 300 K) or semiconductor QW's (~1 nm, even at low temperature), has been until now a major hindrance in this respect, leading to SE enhancement factors of the order of unity in the weak coupling regime [3,10].

Self-assembled InAs QB's constitute an appealing alternate class of light emitters [11]. Owing to their discrete density of electronic states, individual QB's exhibit a single, very narrow emission line (<0.1 nm) under weak excitation conditions [16], which allows implementing high Q and high F_p microcavities. We have inserted such QB's in the core of pillar microresonators, and we report in this Letter the observation of a (up to) fivefold enhancement of their SE when on resonance with a confined cavity

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mode. We show that the magnitude of this effect can be quantitatively explained by considering both the cavity figure of merit F_p and the effect of the random spatial and spectral distributions of the QB's.

The scanning electron micrograph (SEM) shown in Fig. 1 highlights the structure of the micropillars under study. These objects have a circular cross section, a core diameter d in the 40–0.7 μm range, and have been processed [11] through the reactive ion etching (RIE) of a GaAs/AlAs planar microcavity grown by molecular beam epitaxy. Apparent on this SEM is the one-wavelength-thick cavity layer of GaAs, inserted between a 15 period top DBR and a 25 period bottom DBR. Layer thicknesses are designed for the on-axis resonance energy of the planar cavity to be 1.33 eV. Five arrays of InAs QB's (density: 4×10^{10} cm^{-2} per array) have been inserted within the GaAs cavity layer, close to the antinodes of the on-axis resonant mode of the planar cavity. More precisely, two arrays are located at 10 nm from the interface with the top or bottom DBR's, and the other three are, respectively, at the center of the cavity layer, 10 nm above it, and 10 nm below it. In a reference sample without DBR's, the emission of such QB arrays is spectrally broad (60 meV), due to QB size fluctuations, and centered around 1.33 eV.

The combination of the waveguiding by the sidewalls and of the longitudinal confinement by the DBR's allows the pillar to sustain a discrete set of 3D confined modes [9–11]. A simple estimate of their energies and field distributions can be obtained by developing these modes

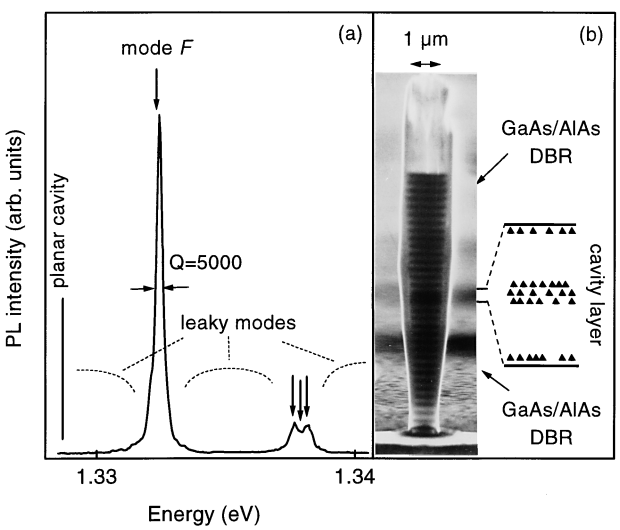

FIG. 1. (a) Typical cw PL spectrum, obtained for a 3 μm diameter micropillar containing InAs QB's. The vertical bar and the arrows indicate, respectively, the resonance energy of the planar cavity prior to etching and the calculated energies for the resonant modes of the pillar. The noise level for this spectrum is 100 times smaller than the background PL from leaky modes. (b) SEM displaying a GaAs/AlAs micropillar; its diameter d, measured in the cavity region, is 1 μm. The location of the five QB arrays within the GaAs cavity layer is sketched in the inset.

as linear combinations of the guided modes of a GaAs cylinder [11]. Focusing our attention on the fundamental resonant mode of the pillar, hereafter labeled F, we can estimate within this framework its effective height ($\sim 2\lambda_c/n$) and area ($\sim \pi d^2/16$). Such a micropillar as shown in Fig. 1 is thus able to confine light within an effective volume as small as $\sim 5\ (\lambda_c/n)^3$.

These confined modes can be conveniently probed by microphotoluminescence (mPL), using the QB arrays as a broadband internal light source [11] (see Fig. 1). These modes contribute a series of sharp lines to the mPL spectra, which allows us to measure their energies and Q's. In addition, we observe for the thinnest pillars a broad featureless background related to the SE from off-resonance QB's into leaky modes. This emission, which exits the pillar through its sidewalls, can be partially collected by our setup for small enough diameters ($d < 4\ \mu$m). Coming back now to mode F, Q is very close to the planar cavity value ($\sim$5200) for diameters larger than 3 μm. For thinner pillars, the scattering by the roughness of the sidewalls reduces the photon lifetime inside the cavity, and thus Q. For the present optimized RIE process, Q is still close to 2000 for mode F for some 1 μm diameter pillars, which corresponds to a record Purcell factor for such microcavities ($F_p = 32$).

We thus expect to observe a clear Purcell effect when studying such small radius pillars containing QB's. Obviously, only those QB's which are both well matched spectrally with mode F and located close to its antinode (i.e., near the center of the pillar) will experience a strong enhancement of their SE rate. In spite of the random spectral and spatial distribution of the QB's, this effect leads, as shown in the following, to a global shortening of the average radiative lifetime of the QB's which emit photons into mode F. Time-resolved mPL is a convenient technique for studying such effects. For low enough temperatures, charge carriers are trapped in QB states. Unlike QW's, their migration toward nonradiative centers is quenched, as shown previously for InAs QB's in a dislocated GaAs layer [17] or placed at 50 nm from a RIE etched surface [16]. The PL decay time τ_d thus reflects accurately the radiative lifetime of trapped electron-hole pairs.

Single pillars have been studied at 8 K, using a setup based on a streak camera and on a Ti:sapphire laser delivering 1.5 ps pulses at 82 MHz. A microscope objective is used to focus the laser beam within a 3 μm diameter laser spot and to collect the PL. For good control of the excitation of the QB system, we use a 1.48 eV laser energy, which is below the band gap of GaAs and above the stop band of the DBR's. Under such conditions, the charge carriers are photogenerated in the InAs wetting layers of the QB arrays, which ensures a good transfer toward confined QB states. The average excitation power P_{ex} lies in the 0.5–5 μW range, which corresponds to an estimated density of photoexcited carriers of the order of 0.1 to 1 electron-hole pair per QB for each laser pulse. Such weak excitation conditions ensure that only the fundamental optical transition of the QB's contributes to the SE, which is essential to get reliable information on the cavity effect.

We compare in Figs. 2(*a*) and 2(*b*) the time-resolved PL spectra obtained for a given class of QB's either placed in bulk GaAs or on resonance with mode F in the pillar shown in Fig. 1(b) ($d = 1\ \mu$m, $F_p = 32$). For this pillar, mode F is centered at 1.351 eV so that we collect for both spectra the PL emission within the 1.351 $\pm$ 0.001 eV spectral window. For the bulk reference, we observe a short PL rise time (<50 ps), which highlights the efficient capture and relaxation of the photogenerated carriers, and a monoexponential decay curve with a 1.3 ns time constant, in agreement with previous experimental work [17]. A much faster decay is observed for QB's on resonance with mode F ($\tau_d^{\text{on}} \sim 250$ ps). As expected in this low excitation regime, the shape of the profile and τ_d do not change significantly with P_{ex} [18]. We consider this clear shortening of τ_d, which is also observed when QB's are on resonance with other confined modes of the pillar, as being due to the Purcell effect.

The study of the SE of off-resonance QB's into the leaky modes provides a firm support for this interpretation. The efficient collection of this SE by our setup for $d < 2\ \mu$m allows the study of τ_d in a single experiment for both on-resonance QB's and QB's detuned by a few meV with respect to mode F. For such a small band gap difference, QB's in bulk GaAs exhibit the same τ_d. The temporal profile shown in Fig. 2(*c*) has been obtained for the same pillar as Fig. 2(*b*) by integrating the signal from off-resonance QB's over the 1.339–1349 eV spectral range in order to improve the signal to noise ratio. In clear contrast with on-resonance QB's, their PL decay rate ($\tau_d^{\text{off}} \sim 1.1 \pm 0.2$ ns) is close to the lifetime of reference QB's in bulk GaAs. The same behavior is observed for off-resonance QB's emitting slightly above mode F. As a result, nonradiative recombination at etched sidewalls is not likely to explain the lifetime shortening observed for on-resonance QB's, which can be unambiguously attributed to a selective SE enhancement due to their resonant coupling with mode F.

This effect has been studied for variable pillar diameters (Fig. 3). For each pillar under study, the on-resonance lifetime of the QB's, τ_d^{on}, is extracted from a monoexponential fit of the PL decay over the first nanosecond after the pulse. Within experimental accuracy, we observe a smooth regular increase of $1/\tau_d^{\text{on}}$ as a function of Purcell's factor F_p. It does not exhibit such a monotonic dependence on Q or V since Q displays large fluctuations from pillar to pillar for a given pillar size in the 1–2 μm range. As expected, the Purcell factor is the relevant microcavity figure of merit for our problem.

For a more quantitative analysis we must also consider, besides F_p, how the emitters are coupled to the vacuum field. First, their electric dipole is randomly oriented in good approximation as shown by recent PL studies [19]. Second, the SE rate of a given QB into a cavity mode depends on its location $\vec{r}$ and emission wavelength λ_e, which govern, respectively, the amplitude of the vacuum field it feels and the density of modes to which it is coupled. Finally, the global SE rate $1/\tau$ of a given QB combines the emission into the leaky modes of the micropillar, $1/\tau_{\text{leak}}$, and the emission into mode F, which is twofold polarization degenerate [9,11]. We assume that $1/\tau_{\text{leak}}$ is close to $0.8/\tau_0$, where τ_0 is the emitter's lifetime in bulk GaAs, as for planar GaAs/AlAs cavities [4,6]. To

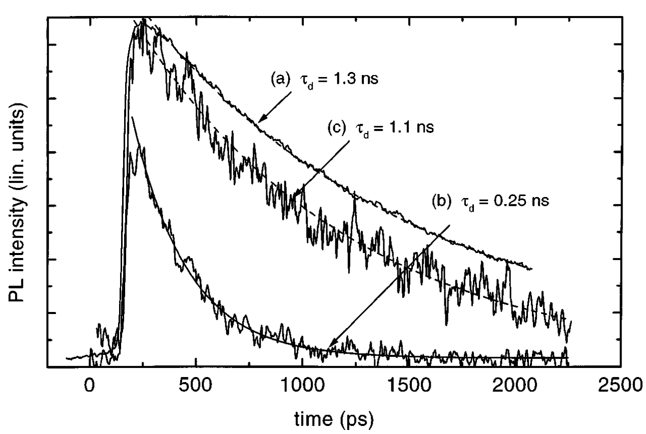

FIG. 2. Time-resolved PL spectra for QB's in a GaAs matrix (*a*) or in the core of the pillar shown in Fig. 1(b) and either placed on resonance (*b*) or out of resonance (*c*) with the fundamental mode. The dashed lines are monoexponential fits of the decay. The solid line in (*b*) features the result of our theoretical model.

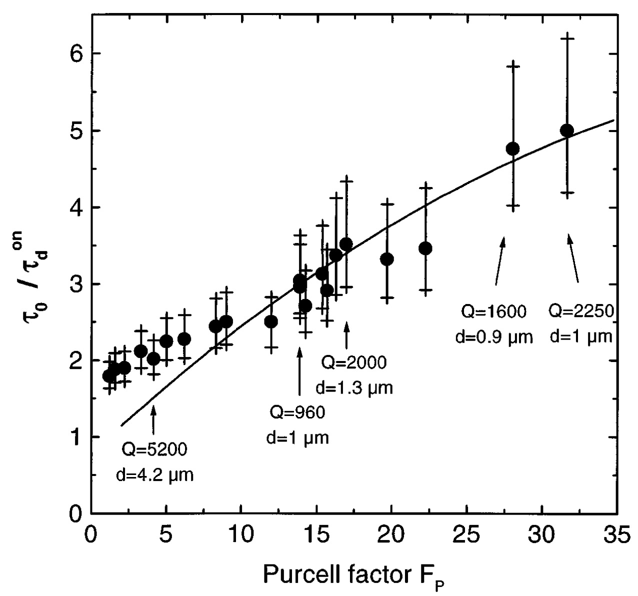

FIG. 3. Experimental (dots) dependence of the PL decay time τ_d^{on} as a function of the Purcell factor F_p. d and Q are indicated for some of the pillars under study. Error bars correspond to a $\pm$70 ps uncertainty on τ_d^{on}. The solid line shows the result of our calculation of the average lifetime of on-resonance QB's.

summarize,

$$\frac{\tau_0}{\tau} = \frac{2F_p}{3} \frac{|\vec{\varepsilon}(\vec{r})|^2}{|\vec{\varepsilon}|^2_{\max}} \frac{\Delta\lambda_c^2}{\Delta\lambda_c^2 + 4(\lambda_e - \lambda_c)^2} + 0.8\,.$$

Even for our thinnest pillars ($d = 0.9\ \mu$m) the number of QB's coupled to mode F is large enough ($\sim$30) for allowing a statistical averaging of the effect of the spatial and spectral distributions. For that purpose, we assume that the QB's are randomly distributed and homogeneously excited during the pulse. The temporal evolution of the emission in mode F is then obtained through a spatial averaging over the pillar cross section and a spectral averaging over the energy window corresponding to the resolution of our experiment ($\lambda_c \pm 1$ nm). As shown in Fig. 2(b), a satisfying agreement with the experimental temporal behavior of the PL is obtained, without any adjustable parameter.

As for experimental profiles, such a theoretical PL decay curve can be reasonably approximated by a single exponential during the first nanosecond after the pulse. We compare in Fig. 3 this calculated initial decay time to our experimental data. Here again, our description of Purcell effect for this inhomogeneous collection of emitters accounts well for the experimental results. We note also a slight systematic deviation for the largest pillars ($F_p < 8$ on Fig. 3). Such structures are wider than our laser spot ($d > 3\ \mu$m), which can lead to a preferential excitation of QB's located close to the center of the pillar, and better coupled to the vacuum field. A detailed discussion of this effect is beyond the scope of this Letter.

Clearly, InAs self-assembled QB's are outstanding light emitters for observing SE enhancement in solid-state microcavities, which opens novel avenues for fundamental physics and optoelectronics. Vertical-cavity light-emitting diodes (LED's) are potential challengers of lasers for, e.g., telecom distribution, owing to their narrow linewidth, reliability, and easy coupling to optical fibers. Using the Purcell effect on a QB active medium might allow us to obtain in a near future pulse decay times in the 50 ps range and LED operation frequencies up 10 GHz, only limited by carrier relaxation dynamics [20]. In principle, using much bigger InAs QB's, emitting, e.g., around 1 eV, should allow us to implement the Purcell effect up to room temperature.

These results also pave the way to fascinating experiments on single QB's in high F_p microcavities. A solid-state replica of the ultimate "one atom + one mode" CQED system can be obtained through the insertion of a very dilute QB array (1 QB per μm^2) in a micropillar with elliptical cross section and nondegenerate fundamental mode [21]. A temperature tuning of the QB emission energy should allow us to demonstrate on a single light emitter the effect of the resonant coupling to a discrete mode. For the present state of the art, the SE enhancement factor would be about 10 on resonance. In this regime, the QB emits only one photon into leaky modes when nine photons are funneled into the cavity mode, which corresponds to an extremely good figure of merit β for the SE coupling efficiency ($\beta \sim 0.9$). Finally, the photon flux in mode F should exhibit as for single atoms a clear antibunching behavior. Owing to this unique set of properties, such solid-state light emitters might open very interesting opportunities for the generation of photon number squeezed states of light and, in particular, for quantum cryptography [3].

The authors are very grateful to the technological team of CNRS-L2M for expert support and to C. Weisbuch, J. Bloch, B. Jusserand, and I. Abram for fruitful conversations. J. M. G. is a member of DGA, French Ministry of Defense.

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