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Enhanced spontaneous emission rate from single InAs quantum dots in a photonic crystal nanocavity at telecom wavelengths

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The authors demonstrate coupling at 1.3 μm between single InAs quantum dots (QDs) and a mode of a two dimensional photonic crystal (PhC) defect cavity with a quality factor of 15 000. By spectrally tuning the cavity mode, they induce coupling with excitonic lines. They perform a time integrated and time-resolved photoluminescence and measure an eightfold increase in the spontaneous emission rate inducing a coupling efficiency of 96%. These measurements indicate the potential of single QDs in PhC cavities as efficient single-photon emitters for fiber-based quantum information processing applications. © 2007 American Institute of Physics.

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The efficient extraction of spontaneous emission from semiconductors still represents a major challenge due to total internal reflection at the semiconductor/air interface. In particular, single-photon sources based on single quantum dots (QDs) are currently plagued by low extraction efficiency and poor coupling to single-mode fibers, typically on the order of 10^{-3} – 10^{-4} , which prevents their application to quantum communication. However, the radiative emission rate and the far field pattern of a two-level system, such as an exciton in a QD, are not intrinsic properties of the emitter itself but are molded by the environment that surrounds it. The local density of states of electromagnetic modes may be shaped in such a way as to increase this emission rate¹ into a given mode of the optical field, which can be extracted efficiently. Photonic crystal (PhC) nanocavities conveniently address this objective by providing modes with the required small volumes and high quality factors. They also allow engineering of the far field pattern of the cavity modes and thus of the collection efficiency.² Increased spontaneous emission rate from single QDs in PhC cavities has been demonstrated at wavelengths smaller than 1050 nm.^{3–7} In this paper, we report on PhC defect nanocavities around 1300 nm with quality factor (Q) as high as 15 000 and their coupling to single excitons, resulting in increased spontaneous emission rate.

The sample, grown by molecular beam epitaxy, consists of a 320-nm-thick GaAs membrane on top of a 1.5 μm $\text{Al}_{0.7}\text{Ga}_{0.3}\text{As}$ sacrificial layer. A single layer of low areal density (5–7 dots/ μm^2) self-assembled InAs QDs emitting at 1.3 μm at low temperature⁸ is embedded in the middle of the membrane to maximize the coupling to the fundamental waveguide mode. The fabrication process of the PhC nanocavities consists in e-beam patterning of a polymethyl methacrylate resist and transfer to a 150-nm-thick SiO_2 layer by

CHF_3 plasma etching. This serves as a mask for the pattern transfer onto the GaAs layer by $\text{SiCl}_4/\text{O}_2/\text{Ar}$ reactive ion etching. The sacrificial layer is then selectively etched in a diluted HF solution to release the GaAs membrane. As a result, we obtain a PhC structure with seven shells of holes around the nanocavity at the center of a 12 μm diameter suspended membrane. The $L3$ defect cavity presented hereafter (see the inset of Fig. 1) is formed by keeping three holes unetched along the ΓK direction of the triangular PhC and parallel to a cleaved edge of the GaAs sample. The lattice constant $a=340$ nm and circular holes with diameter $d=125$ nm provide a photonic band gap for TE polarization around 1.3 μm . This corresponds to the ground state emission wavelength of our QDs at ~ 10 K. To obtain a smoother mode field pattern, and thus a higher Q , we modified the first holes on both ends of the cavity.⁹ They are shifted by $a'=0.15a$ outwards, and their diameter is $d'=2/3d$, following experimental optimization.¹⁰

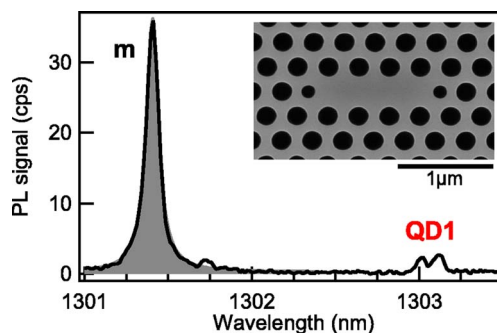


FIG. 1. (Color online) μPL spectrum of the modified $L3$ cavity around its fundamental mode (m) under pulsed excitation (80 MHz, 80 nW, and 750 nm). A Lorentzian fit (gray area) of the mode yields a quality factor $Q=14\,850$. A doublet excitonic line labeled QD1 is visible at lower energies. The inset shows a scanning electron microscopy image of this cavity.

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The sample was kept in a liquid He continuous flow cryostat and all measurements were performed at ~ 10 K. We used a pulsed laser (pulse width of ~ 50 ps) at 750 nm for excitation in the GaAs. The microphotoluminescence (μ PL) signal is collected through the same microscope objective (numerical aperture=0.5) and then coupled to a single-mode fiber. The fiber acts as a pinhole and ensures a small collection area on the order of $2 \mu\text{m}^2$. It can be connected to either a single-photon detector for time correlated single-photon counting or to an $f=1$ m spectrometer equipped with a LN_2 -cooled InGaAs array providing a spectral resolution of $\sim 34 \mu\text{eV}$. Figure 1 shows a typical μ PL spectrum obtained at an average excitation power of ~ 80 nW. Interestingly, the cavity mode is visible, even when it is spectrally far from excitonic lines. We attribute this to an energetically broad multiexcitonic background emission correlated to the QD, but whose exact nature is still not clearly understood.^{7,11} The cavity mode has a Lorentzian lineshape. Its full width at half maximum of $64 \mu\text{eV}$ yields a quality factor $Q=E/\Delta E=\omega_c/\Delta\omega_c \approx 14\,850$, which is an indication for the high quality of the sample processing. This is also promising for the control of spontaneous emission, since this corresponds to a maximum achievable Purcell factor $f_P=(3/4\pi^2)(\lambda_c/n)^3(Q/V_m) \approx 1500$, assuming⁹ a mode volume $V_m \approx 0.73(\lambda_c/n)^3$. As the position of the emitter is random and, in general, not perfectly matched to the cavity field, we expect to operate in the weak-coupling regime with a lower Purcell factor.

A doublet emission line labeled QD1 is visible at 1303.0 nm. Based on temperature- and power-dependent measurements, these lines can be definitely ruled out as cavity mode. μ PL as well as time-resolved measurements indicate that both peaks originate from the same spatial location and we attribute this doublet structure to the splitting of the bright exciton angular momentum states due to QD asymmetry.¹² The cavity mode, whose polarization varies spatially, likely couples to both dipoles of the splitted exciton.

Temperature tuning is commonly used to spectrally bring the QDs in resonance with the cavity since they redshift faster than the mode.⁶ However, the temperature increase changes the relative intensity of the various excitonic lines of a single QD and causes line broadening,⁸ making the analysis of QD-cavity coupling difficult. Moreover, since these particular QD lines are already on the red side of the mode, we need another tuning strategy. We noticed that the cavity mode naturally redshifts during the measurement at a rate of a few tenths of nm/h before reaching an equilibrium position. Upon heating the cryostat to room temperature and subsequent cooling, the mode is reset at higher energy. We attribute this reversible shift to residual gas molecules adsorbed on the sample surface, as in cryopumping systems, which decrease the filling factor of the PhC and modify the resonant energy of the mode.¹³ Spectra taken at different times clearly show the red-shift of the mode and its crossing over the QD excitonic lines [Fig. 2(a)]. As the energy detuning between the mode and the QDs decreases, we observe an enhancement of the integrated μ PL signal from QD1. This rather clearly indicates coupling with the cavity mode: photons emitted by the exciton efficiently escape the GaAs through the mode emission pattern. No splitting is observed at resonance, indicating that despite the high Q value, the QD-cavity system is not in the strong-coupling regime, prob-

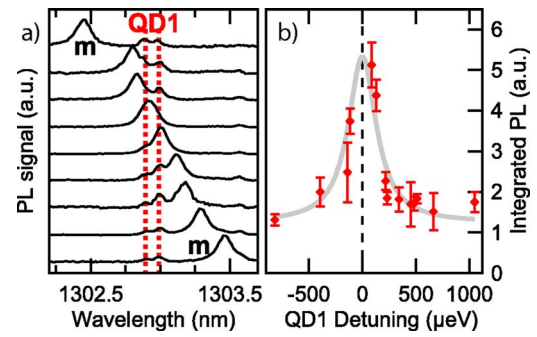


FIG. 2. (Color online) (a) μ PL spectra (15 nW, 80 MHz) showing a QD (1303.0 nm) and the redshift of the mode (m) due to air molecules being cold trapped on the surface of the sample. As the mode approaches QD1, we observe a marked increase in its emission intensity. (b) Integrated intensity of QD1 as a function of the detuning with respect to the mode energy.

ably due to nonoptimal spatial coupling.¹⁴ The integrated intensity of the exciton as a function of the detuning [Fig. 2(b)] was obtained by fitting the region of interest in the μ PL spectra with the sum of a Lorentzian for the mode and two Lorentzians for QD1. It follows a Lorentzian trend, as expected in the case of Purcell enhancement, with a width proportional to τ_0/τ .

To confirm that this emission enhancement is due to the Purcell effect and to quantify it, we performed time-resolved measurements. The μ PL signal is spectrally filtered by a tunable filter with 0.8 nm bandwidth and directed to a fiber-coupled superconducting single-photon detector (SSPD).¹⁵ The histogram of delays between the laser pulse and the detector output is recorded. The SSPD has a quantum efficiency of 10% and a dark count rate of 8 Hz at a bias current of 21 μA . It provides a much higher sensitivity and signal to noise ratio than InGaAs avalanche photodiodes.¹⁶ The temporal resolution of the system is around 150 ps and deconvolution was performed to extract the emitter lifetime. The sample was excited with ~ 5 nW average power at 20 MHz. The measured lifetime of QDs away from the cavity (Fig. 3, diamonds) is $\tau_{\text{QD}} \approx 1.2$ ns. This is in good agreement with previously reported values for the exciton lifetime in single QDs grown under the same conditions.¹⁷ For QD1, in the cavity, but energetically off resonance, we observe a suppression of the spontaneous emission rate, as shown in Fig. 3 (empty circles), where for a 2.5 nm detuning, the lifetime of QD1 is $\tau_{\text{off}}^{\text{QD1}} \approx 3.6$ ns. In fact, when the cavity mode is off

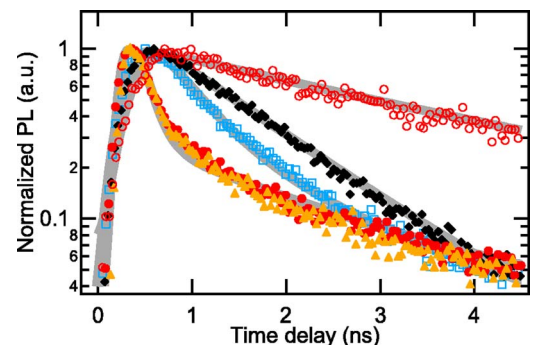


FIG. 3. (Color online) Time resolved dynamics of QD1 off resonance (empty circles), of ensemble QDs outside the PhC region (diamonds), of the background emission coupled to the mode off resonance with QD1 (empty squares), and of QD1 on resonance with the cavity mode under 5 nW (filled circles) and 0.4 nW (filled triangles) excitation powers and a 20 MHz repetition rate.

the QD line, the density of states of electromagnetic modes at the QD energy is smaller than in free space resulting in an inhibition of the spontaneous emission and thus a slower decay rate through the leaky modes, as shown already.^{5,6} The PL decay time of the QD on resonance (Fig. 3, filled circles) clearly shows two time constants. The fast one, $\tau_1^{\text{QD1}} \approx 150$ ps, is attributed to the Purcell effect and indicates a $(\tau_{\text{QD}}/\tau_1^{\text{QD1}}) \approx$ eightfold rate enhancement. The slow time constant, $\tau_2^{\text{QD1}} \approx 1.9$ ns, is attributed to the spin flip transition from dark to bright exciton⁵ or the spectrally broad background emission coupled into the filter bandwidth. As both the exciton and the background couple to the cavity line in the resonance condition, and since the background emission presents a stronger power dependence than the QD1 line, we used a low pump power (5 nW) such that the count rate of the mode off resonance is more than four times lower than QD1 on resonance. This ensures that most of the signal is indeed originating from the QD1 line. Moreover, at a much lower power of 0.4 nW (Fig. 3, triangles), where the background is further suppressed with respect to the exciton, the same decay curve is obtained. In contrast, the background emission coupled to the mode off resonance with QD1 presents a different decay rate (Fig. 3, squares) with a deconvoluted lifetime of 370 ps. The coupling efficiency into the mode defined⁵ as $\beta = 1 - (\tau_1^{\text{QD1}}/\tau_{\text{off}}^{\text{QD1}})$ is $\sim 96\%$. This result is very promising; as most photons are emitted into the cavity mode, a very large collection efficiency can be obtained by optimizing the far field of this mode.²

In summary, we presented modified *L3* defect nanocavities on GaAs with very high *Q* of 15 000 at 1300 nm wavelength. We observed a clear enhancement of μ PL signal of single excitons as a function of the energy detuning to the cavity mode. We further demonstrated Purcell effect on this cavity by time-resolved analysis of the dynamics of two neighboring transitions at 1.3 μm . We obtained an eightfold enhancement of spontaneous emission rate yielding a $\beta \approx 96\%$ coupling efficiency. The reduced QD lifetime may also lead to coherent, transform-limited single-photon sources. Together with the increased extraction efficiency, it pledges the realization of efficient PhC nanocavity based single-photon emitters at 1.3 μm for optical quantum information processing.

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