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RAPID COMMUNICATIONS

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Controlling the charge environment of single quantum dots in a photonic-crystal cavity

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We demonstrate that the presence of charges around a semiconductor quantum dot (QD) strongly affects its optical properties and produces nonresonant coupling to the modes of a microcavity. We show that, besides (multi)exciton lines, a QD generates a spectrally broad emission which efficiently couples to cavity modes. Its temporal dynamics shows that it is related to the Coulomb interaction between the QD (multi)excitons and carriers in the adjacent wetting layer. This mechanism is suppressed by the application of an electric field, making the QD closer to an ideal two-level system.

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The study of single quantum dots (QDs) embedded inside photonic-crystal (PhC) cavities or micropillars has been the subject of an intense interest for both fundamental science and applications.[1] Indeed, the discrete energy structure in a QD makes it a semiconductor equivalent of an atomic system and allows solid-state implementations of cavity quantum electrodynamic experiments, with applications to single-photon sources, entanglement generation, and quantum computing. However, the microphotoluminescence (micro-PL) experiments performed on QDs coupled to optical microcavities have revealed a phenomenon contradicting the ideal atom-in-a-cavity model: the cavity mode emission was observed despite the lack of an excitonic transition in resonance with the mode.[2–13] As the cavity emission was observed to provide a classical photon statistics,[2] this behavior undermines applications of QDs to quantum information processing, for example, reducing the purity of single-photon sources. So far, several physical mechanisms have been evoked to explain this observation: dephasing processes,[6–8,10] a continuum in the hole states,[4] and a continuum due to a mixing between *s* and *p* states.[3] Experimental results have provided evidence of the role of dephasing in nonresonant coupling to cavity modes in the case of small detuning (1–3 meV).[5,11–13] In this Rapid Communication, we show the existence of a second process which provides nonresonant coupling at much larger detunings (up to 10 meV). It originates from the Coulomb interaction between the carriers in the wetting layer (WL) and the multiexcitons in the QD, which generates a spectrally wide emission coupled to the mode. We support this interpretation by investigating the dynamics of the QD-cavity system. Additionally, we show that the charge environment around the QD can be controlled by the application of an electric field, which brings our system closer to an ideal two-level system.

We investigate InAs self-assembled QDs in PhC cavities, although we expect that our conclusions also apply to other types of cavities, such as micropillars and microdisks. A typical low-temperature (5 K) emission spectrum from a L3 cavity (three missing holes[14]), as measured in a microphotoluminescence setup, is shown in the inset of Fig. 1. The sample, grown by molecular beam epitaxy, consists of a 320-nm-thick GaAs membrane on top of a 1.5 μm $Al_{0.7}Ga_{0.3}As$ sacrificial layer. A single layer of low-density ($5-7$ dots/μm^2) self-assembled InAs QDs emitting at 1.3 μm at low temperature is embedded in the middle of the membrane.[14]

A strong cavity mode emission with a quality factor of 11 500 and two sharp lines associated to single QDs are observed (inset of Fig. 1). The PL decay was measured by time-correlated fluorescence spectroscopy using a gain-switched 750 nm pump laser, a fiber-based tunable filter, and a superconducting single-photon detector,[15] providing combined spectral and temporal resolutions of 0.8 nm and 150 ps, respectively (Fig. 1). The excitonic line QD1 (QD2) has a monoexponential decay with a lifetime of 2.6 ns (3.1 ns), longer than the intrinsic radiative time of 1.1 ns,[16] due to the low available optical density of states in the off-resonant cavity. In contrast, the cavity mode emission has a biexponential decay with a fast lifetime of 0.4 ns and a slow lifetime of 1.3 ns. 85% of the cavity mode emission comes from the fast decay which is clearly distinct from the decay of the excitonic lines QD1 and QD2. This behavior, typically observed in our samples for all the investigated detunings (2–10 nm) and already reported in Ref. 14, shows that in these structures the cavity mode emission cannot be due to the homogeneous broadening of the excitonic lines. Indeed,

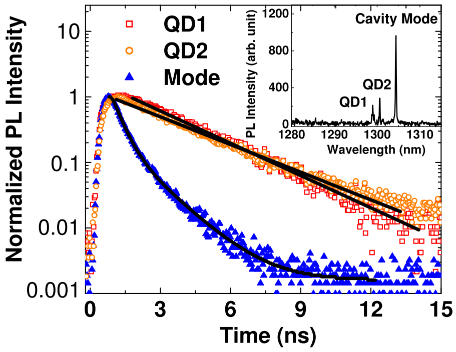

FIG. 1. (Color online) Time resolved experiments of the cavity mode and off-resonance QDs. Inset: spectrum of a L3 PhC cavity, under pulsed excitation ($\lambda = 750$ nm, average pump power $P_{av} = 0.4$ μW in a 2 μm diameter spot), showing the cavity mode emission and two off-resonance excitonic lines from QDs. The fits of the experimental decays are shown in black.

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in the latter case both lines would mirror the decay of a single emitter resulting in the same dynamics.[12]

We instead propose that the off-resonant cavity peak is pumped by a spectrally wide emission (to be referred to as "background") due to radiative recombination of QD (multi)excitons, dressed by the Coulomb interactions with carriers in the wetting layer. Such emission, also previously reported,[17–20] is strongly enhanced by the cavity coupling, as also confirmed by the fast lifetime observed in Fig. 1(b). In order to gain more insight in the carrier dynamics within a QD, we have investigated the PL decay of a QD in the absence of a cavity. To this aim a layer of low-density QDs was grown in a $\lambda/2$ planar cavity,[21] providing increased coupling into the collection objective with negligible alteration of the spontaneous emission rate. Small (1 μm^2) apertures in a gold mask were processed to isolate a single QD. Figure 2(a) shows the time-integrated spectrum of a single QD under a $P_{\mathrm{av}}=6$ μW excitation at 6 K. Single lines associated to neutral or charged multiexcitonic states are clearly observed together with a broad background emission, which extends over >10 nm, depending on the excitation density. Figure 2(b) shows a streak-camera-like image of the PL decay at different wavelengths, obtained by scanning a tunable fiber bandpass filter (full width at half maximum of 0.3 nm) and measuring the decay at each wavelength by time-correlated fluorescence spectroscopy.

In the first phase of the decay (<1 ns), a continuous emission and featureless is observed from 1286 to 1308 nm. Then, from 1 to 3 ns, the emission is still broad but the intensity of the emission is stronger around 1295 nm where the majority of the single lines are observed. After 3 ns, the background emission progressively disappears and, at the end, the QD emission comes mainly from the single lines of the QD. The decays at two different wavelengths, corresponding to a single line (1295.5 nm), attributed to a biexciton[22] and to the featureless background (1306.5 nm), are also shown in Fig. 2(c) in a logarithmic scale. The biexciton emission is clearly delayed as compared with the background: the maximum of intensity is reached after 1.2 ns for the background emission and 2 ns for the biexciton. The delayed biexciton emission suggests that (multi)excitonic lines take place after recombination of carriers in the two-dimensional WL continuum, which supports our interpretation of the origin of the background emission. This behavior also agrees with the anticorrelation between excitonic line and cavity mode observed in Ref. 2. Moreover, as shown in Fig. 2(c), the decay of the cavity mode studied in Fig. 1 under a $P_{\mathrm{av}}=4$ μW excitation clearly reproduces the background decay. This confirms the fact that the cavity mode is pumped by the background and indicates that the biexponential decay of the mode is due to a biexponential decay of the background, whose origin is not clear. As shown on Fig. 2, the background emission is a complex phenomenon, with a frequency-dependent decay dynamics. Indeed, the different energies within the broad emission are connected to different charge densities and configurations of the carriers surrounding the QD. The temporal decay of this carrier population leads to a reshaping of the background emission as a function of time, ultimately resulting in clean (multi)excitonic lines.

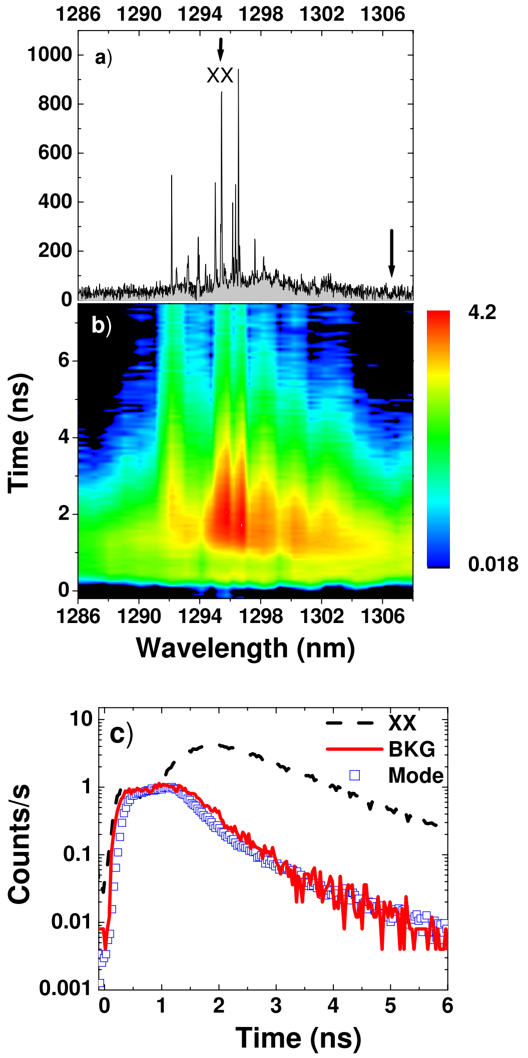

FIG. 2. (Color online) (a) Spectrum of a single QD and (b) time-resolved experiments performed on the same QD. (c) Two time-resolved PL decays from (b), corresponding to [see arrows in (a)] the biexciton line (black dashed line) at 1295.5 nm and background emission at 1306.5 nm (red line). The measurement of the cavity mode studied in Fig. 1 under a 4 μW excitation is also shown (blue squares). The intensity of the cavity mode has been normalized to fit the background emission.

In order to further support our interpretation, we investigate the effect of an electric field on the emission spectrum in a PhC diode structure under reverse bias. The diode consists of a 370-nm-thick GaAs/AlGaAs heterostructure with *p*- and *n*-contact layers on the two sides, incorporating a single layer of low-density InAs QDs. The fabrication process is described in Ref. 23.

The photoluminescence of a L3 PhC diode has been studied using a cw 660 nm laser ($P_{\mathrm{av}}=5$ μW) under an applied voltage. PL spectra are presented in Fig. 3(b) as a function of the applied voltage V_b (defined as positive in reverse bias). For a small reverse bias (0–0.5 V) a cavity mode is observed at 1290 nm with a quality factor of 850 along with a wide and unstructured background emission. A strong modification of the spectrum is observed when a bias voltage >1 V is applied: the cavity mode and the broad background disap-

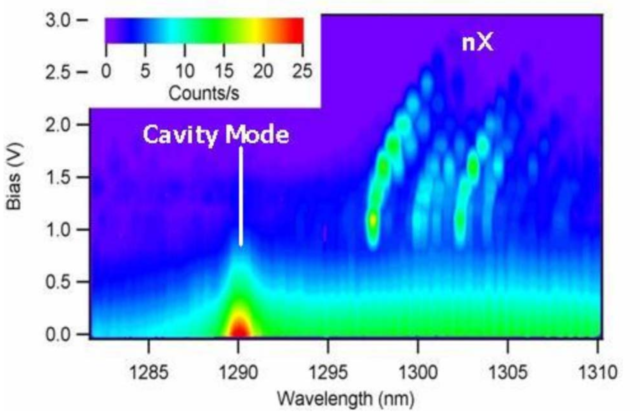

FIG. 3. (Color online) Observation of the ground state emission of the PhC diode as a function of the bias voltage.

pear and (multi)excitonic lines ("nX") are observed. Our attribution of cavity and excitonic lines is confirmed by the field dependence of their energy. Indeed, while the cavity energy does not vary with the voltage, the excitonic lines show a large Stark shift,[24] corresponding to a dipole moment $p = 1.1 \times 10^{-28}$ C m and polarizability $\beta = -4 \times 10^{-36}$ C m^2 V^{-1}, close to values typically observed for InAs/GaAs QDs.[25–27]

The intriguing observation of the disappearance of the cavity mode with applied field and the simultaneous appearance of excitonic lines clearly indicates that the cavity peak is associated to less confined higher-energy carriers, which are more easily swept away by the electric field than confined carriers in the QD. The observed electric-field dependence confirms our interpretation of the cavity emission and, at the same time, provides a means of controlling the QD charge environment and thus retrieving the ideal atom-cavity coupling.

In order to confirm that these additional carriers are indeed located in the WL, we have studied the WL and cavity emission as a function of the electric field. The inset of Fig. 4 shows the micro-PL of the PhC diode over a wide spectral range for two different voltages. At 0 V, two cavity modes are observed: the studied mode emitting at 1290 nm and a second one at 1185 nm. Additionally a strong emission of the WL is observed at 940 nm consistent with previous observations on similar QDs.[28] In contrast, with a 1 V applied bias both the WL and cavity peaks disappear, and single excitonic lines emerge. The integrated intensities of the wetting layer and of the cavity mode at 1.29 μm, reported in Fig. 4, show the same dependence on the electric field confirming that they are both related to the same population. If we assume that the dynamics of the carriers in the wetting layer is driven by a carrier lifetime τ_0 (including all field-independent radiative, nonradiative, and capture processes) and a tunneling channel with a field-dependent time constant $\tau_T(F)$, the intensity of the WL peak as a function of the electric field F is given by $I(F) = I_0/[1 + \tau_0/\tau_T(F)]$. In our case, the tunneling channel $\tau_T(F)$ is the tunneling rate through a triangular barrier [inset of Fig. 4(b)] which equals[29]

$$\tau_T^{-1}(F) = \frac{eF}{4\sqrt{2m^* V_{barrier}}} \exp\left(-\frac{4\sqrt{2m^*}}{3e\hbar F} V_{barrier}^{3/2}\right).$$

The tunneling rate is mainly related to the escape of electrons due to their smaller effective mass as compared to the heavy holes. The experimental results are fitted using a single fitting parameter V_b and fixing $m^* = 0.063 m_0$ (with m_0 as the electron mass) and $\tau_0 = 400$ ps,[28] providing a value $V_b = 135$ meV. The energy gap discontinuity between the wetting layer and the bulk GaAs being equal to 200 meV, we can conclude that $V_{barrier}$ is the energy spacing between the WL electron ground state and the GaAs conduction band edge. We note that the measured temporal dynamics (not shown) of excitonic and cavity lines in the diode structure closely match those of standard PhC cavities (Fig. 1), showing that the findings of Figs. 3 and 4, though obtained in more complex structures, are representative of PhC cavities in general.

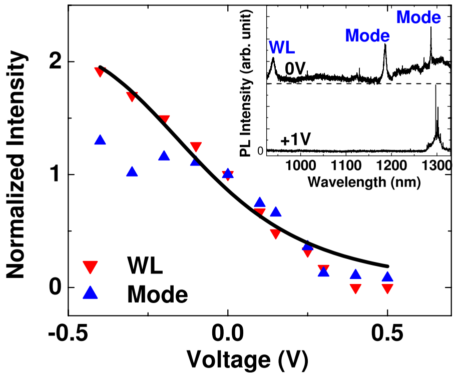

FIG. 4. (Color online) Evolution of the wetting layer and the cavity mode as a function of the reverse bias. Inset: emission of the PhC cavity for $V_b = 0$ and $V_b = 1$ V.

Due to the multiexcitonic nature of the background emission, a cavity mode pumped by the background should behave like a classical emitter. The autocorrelation experiments performed on cavity modes indeed reveal a classical emission[2] under off-resonance excitation conditions similar to those used in our experiments (excitation wavelengths of 750 and 660 nm). In contrast, single-photon emission from the cavity peak has been observed for quasiresonant excitation.[5,11,13] In the latter case, few carriers are created in the WL and the cavity mode is only pumped by the dephasing processes of a single exciton line, resulting in a nonclassical statistics.

In conclusion, the investigation of the dynamics of a QD-cavity system and of its dependence on the electric field has provided strong evidence that the a nonresonant cavity mode can be pumped by a broad QD emission originating from the Coulomb interaction between confined QD excitons and free carriers in the wetting layer. The application of an electric field removes the WL carriers and therefore brings the QD closer to an ideal two-level system. This mechanism for nonresonant coupling, observed here for relatively large detuning, does not exclude the existence of other phenomena such as dephasing processes already observed for small detuning.

After the preparation of this Rapid Communication, Winger *et al.*[30] reported a theoretical model, which provides a quantitative description of the coupling of QD excitons to WL carriers. Our data provide a direct experimental confirmation of the main assumption in the model of Winger *et al.*

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