Controlling the properties of single photon emitters via the Purcell effect

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Single photon emission by an InAs/GaAs quantum dot weakly coupled to a photonic crystal microcavity has been studied as a function of energy detuning. Precise and continuous control of the photon statistics as well as of the linear polarization emission angle is achieved simply by changing the energy detuning between the exciton and the cavity mode. A continuous decrease of the antibunching time, the bunching amplitude and the $g^2(0)$ value is observed as the detuning is decreased at constant excitation rate, due to the detuning-dependent Purcell effect.

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I. INTRODUCTION

The development of efficient single photon sources for future applications in quantum information handling is a very active research field, as deterministic sources of single photons are considered a crucial prerequisite for the implementation of quantum information processing,[1] particularly for quantum cryptography.[2] Various schemes for single-photon generation[3] have been proposed, including two-photon down conversion using nonlinear crystals,[4] photon blockade[5] and fluorescence of single molecules,[6] trapped ions and atoms,[7] carbon nanotubes,[8] as well as diamond color centers.[9] Due to their versatility, scalability, and ease to handle, single semiconductor quantum dots (QDs) are amongst the most promising candidates of stable, solid-state emitters for such applications.[10]

Control of their properties, such as polarization, emission rate, and timing, is essential for the efficient information exchange between photons and static q-bits, e.g. electron or exciton spins in semiconductor quantum dots.[11] This control can in principle be achieved by coupling the QD excitons to confined optical modes in microcavities (MC)[12] or to plasmons in metal nanostructures.[13] In particular QD-MC coupled systems reveal drastic changes on photon statistics[14] by exploiting the Purcell effect,[15] to enhance the spontaneous emission (SE) rate into a specific cavity mode and therefore achieve fast and efficient single photon emission. Most of the research activity focuses on the strong coupling regime between the QD and the cavity mode, in spite of the difficulty to obtain QDs that are spatially and spectrally matched to the cavity modes. In this regime, coupling enhances the quantum efficiency[16] and reduces the timing jitter.[17] However, under weak coupling the Purcell effect enhances the nonclassical properties of light, and control of the polarization properties of the QD is also possible.[18] It allows therefore investigation of the fundamental properties of single photon emitters as well as control to some extent over their operation.[19]

In this paper, we report on the simultaneous and continuous control of the antibunching time, the bunching amplitude, and the $g^2(0)$ value, together with the angle of the linear polarization (previously studied in Ref. 18) of the light emitted by a single InAs QD weakly coupled to a photonic crystal microcavity (PCM). The coupling provokes a detuning-dependent decrease of the antibunching time due to the Purcell effect, instead of the more common mechanism of increasing the pumping rate. The present results are relevant for the use of weakly coupled QD-cavity systems for quantum information applications, as they constitute an experimental step towards the realization of a single photon source with detuning controlled emission properties on demand.

II. SAMPLE AND EXPERIMENT

The measurements included in this work were performed on a self-assembled InAs QD, located about 900 nm from the center of the cavity. The QD has a ringlike shape[20] and the H1 calzone PCM[21] has a quality factor of about 4000.[18] The lowest energy cavity mode is split into two counterlinearly polarized components separated by 3.2 meV, designated as CMX and CMY modes (where X corresponds to the long cavity diagonal).

Photoluminescence (PL) spectra were collected with a micro-PL setup, where excitation and collection occurred through an objective of $NA = 0.5$, with a Gaussian spot of 1.5 μm half width. The spectrometer slit was adjusted to admit the whole width of the QD emission. In this way, we avoid unwanted effects of spectral diffusion. A Ti-sapphire continuous laser was used for excitation resonant with the QD p states. The excitation intensity was kept constant at 280 μW throughout the experiment, and the resolution of the spectrometer was 100 μeV. Computer-controlled step motors were used to vary the position of the excitation spot with 14-nm steps. This allows us to identify the position of the QD with an accuracy of 300 nm, as a 10-step displacement provokes a 10% decrease in the PL intensity maximum. The only parameter varied in the present measurements was the detuning δ of the QD excitons from the cavity mode ($\delta = E_{QD} - E_{CM}$) by *in situ* Xe thin film deposition[22] at fixed temperature (39 K) and input power. A Hanbury–Brown and Twiss (HBT) interferometer was used for the photon correlation measurements, with a pair of avalanche photodiodes for coincidence detection with 30% efficiency at the QD emission energy and a response time of 0.35 ns. The overall HBT instrument response time $\tau_{IRF} =$ 0.5 ns has been obtained using a picoseconds pulsed laser. Typical values of the integration time and count rate are 5 h and 5000 cps, respectively.

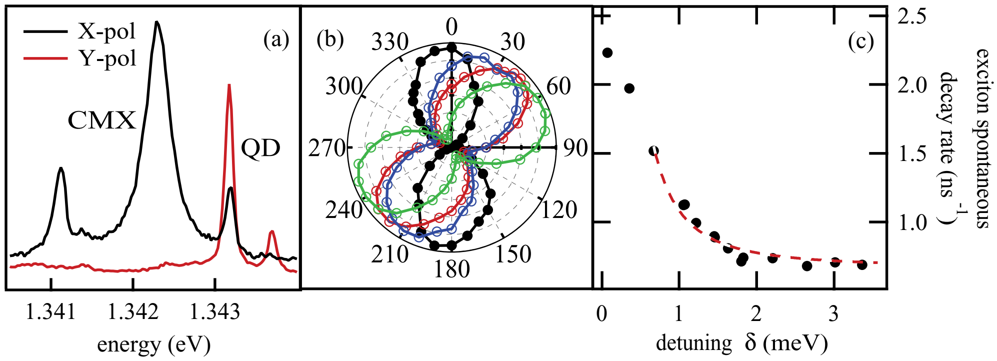

FIG. 1. (Color online) (a) PL spectra of the QD and CM for X (black) and Y (red) polarizations for $\delta = 1$ meV and temperature $T = 39$ K. (b) Polarization polar plots (black dots for the CM and colored hollow dots for the QD) for $\delta = 0.4$ meV (blue [medium gray]), $\delta = 1$ meV (red [dark gray]), and $\delta = 2$ meV (green [gray]). The data are normalized to unity, and the polarization angle $\varphi = 0^\circ(90^\circ)$ corresponds to X(Y) polarization. (c) Exciton spontaneous decay rate, as a function of detuning, under pulsed excitation. The dashed line is a fit using (1).

The PL spectrum of the QD is shown in Fig. 1(a). Distinct emission peaks of the QD and the cavity mode (CMX) for X (black) and Y (red) polarization are observed at 1.3432 and 1.3422 eV, respectively, where X corresponds to the long cavity diagonal. Emission around 1.341 eV originates from another dot nearby, also weakly coupled to the cavity mode.[23] The studied QD can be excited only with Y-polarized resonant excitation. This peculiar behavior can be attributed to the large geometrical asymmetry presented often by these kinds of ring-shaped QDs. The QD linear polarization emission rotates continuously with the energy detuning, as reported in Ref. 18 [Fig. 1(b)] due to hybridization of the QD exciton with the cavity mode. The exciton spontaneous decay rate γ [measured using a pulsed diode laser (866 nm, 60 ps) and one of the HBT detectors] increases as the detuning is reduced [Fig. 1(c)] due to the Purcell effect. Its detuning dependence can be given by[24]

$$\gamma(\delta) = \frac{1}{\tau_0} + \frac{4g^2}{\kappa}\frac{\kappa^2}{4\delta^2 + \kappa^2}, \qquad (1)$$

where τ_0 is the exciton decay time in the absence of coupling, κ stands for the cavity line width and g is the coupling strength between the QD and the cavity mode. From Fig. 1(a), one has $\kappa = 350\,\mu$eV, and from the fit in Fig. 1(c) using $\tau_0 = 1.66$ ns, we obtain $g = 75\,\mu$eV. The spontaneous emission rate of the QD varies from 2.2 ns^{-1} at almost zero detuning to 0.6 ns^{-1} for 2 meV detuning, corresponding to an almost fourfold enhancement of the SE rate due to the Purcell effect. This enhancement can be compared with the Purcell factor $F_P = \frac{3Q(\lambda/n)^3}{4\pi^2 V_m}$. For our sample, the mode volume is $V_m \approx (\lambda/n)^3$ and together with $Q \approx 4000$, they give a Purcell factor $F_P \approx 300$. The large difference between the two values can be attributed to spatial, spectral, and polarization mismatch between the QD and the cavity mode.[25]

III. RESULTS AND DISCUSSION

In order to investigate the effect of detuning on the single photon emission properties of the dot, we perform photon autocorrelation measurements for each detuning at its polarization angle. By Xe deposition, only the range of positive δ is accessible. Negative detunings require temperatures above 50 K, where increased background prevents single photon emission. The results can be seen in Fig. 2, where the second-order correlation function $g^2(\tau)$ is presented for various δ values. At longer delays (measured up to 250 ns) $g^2(\tau)$ tends to 1 for all measurements. Antibunching at zero delay as well as bunching in a longer timescale can be observed. The data

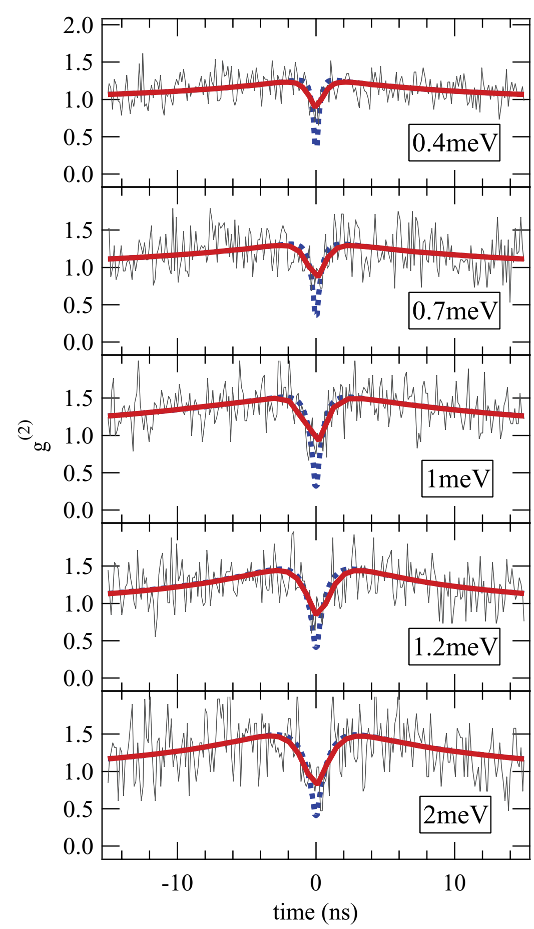

FIG. 2. (Color online) Second order autocorrelation function of the QD for various δ (gray). The data are fitted by Eq. (3) convoluted with the detectors' response function (solid red line). The dotted blue curve represents the unconvoluted function.

in Fig. 2 could be fitted using the formula:

$$g^{(2)}(\tau) = 1 - (1+\alpha)\cdot \exp\left(-{|\tau|}/{\tau_R}\right) + \alpha \cdot \exp\left(-{|\tau|}/{\tau_D}\right). \quad (2)$$

where τ_R and τ_D represent the antibunching and bunching characteristic times, respectively. This expression has been first applied to dye molecules.[26] In order to account for the background emission, that gives a nonzero $g^{(2)}(0)$ value, we modify Eq. (2) as follows:

$$g^{(2)}(\tau) = 1 - \beta \cdot \exp\left(-{|\tau|}/{\tau_R}\right) + \alpha \cdot \exp\left(-{|\tau|}/{\tau_D}\right), \quad (3)$$

where $\beta(\beta \leqslant 1+\alpha)$ and α represent the antibunching and bunching amplitudes, respectively. The red solid traces in Fig. 2 are fits of the experimental data with Eq. (2), convoluted with the instrument response function:[27]

$$h(\tau) = C \cdot \exp\left(-{|\tau|}/{\tau_{\mathrm{IRF}}}\right), \quad (4)$$

with $\tau_{\mathrm{IRF}} = 0.5$ ns. The blue-dashed lines stand for $g^2(\tau)$ without this convolution, i.e. for ideal detectors with zero response time.

The parameter values extracted from the fits in Fig. 2 are presented in Fig. 3 as a function of detuning (lower scale) and corresponding emission polarization angle (upper scale). The $g^2(0)$ values [Fig. 3(a)] decrease with decreasing detuning and lie below 0.5 in the whole detuning range, revealing single photon emission at all detunings below 2 meV (and all polarization angles). The fact that the raw data (which includes convolution) show the opposite trend is due to the instrument response time [Eq. (4)], which becomes comparable with the antibunching time (see below) for the studied detuning range. Intuitively, one would expect that cavity photons will increase $g^{(2)}(0)$ at decreasing detuning. However, this effect is small in our case because the cavity emission intensity at the QD energy is low for the detuning range studied. In fact, background emission composed by cavity photons together with photons of different origin is between 5% and 10% of the QD emission. In our case, the effect of cavity photons is overcompensated by the Purcell effect: as detuning decreases, the QD emission rate increases faster than the background photon emission. Therefore, for moderately small decreasing detunings ($0.4 < \delta < 2$ meV), the probability to collect QD photons increases compared to background photons. In the limit, if the background photon emission is much weaker than QD emission, the value $g^{(2)}(0) = 0$ should be ideally reached. A similar behavior has been reported[19,28] where the relative weight of the background recombination contribution to the cavity emission decreases with decreasing detuning.

The antibunching time τ_R and the bunching amplitude α obtained from Fig. 2 can be interpreted in terms of a metastable state together with the change in the spontaneous emission rate due to the Purcell effect. Their trend with detuning can be modeled following the model of Ref. 26, with the transition rates shown schematically in Fig. 4. The antibunching time τ_R [Fig. 3(b)] shows a continuous decrease for decreasing detuning. The solid line is a fit of the expression for τ_R[26]

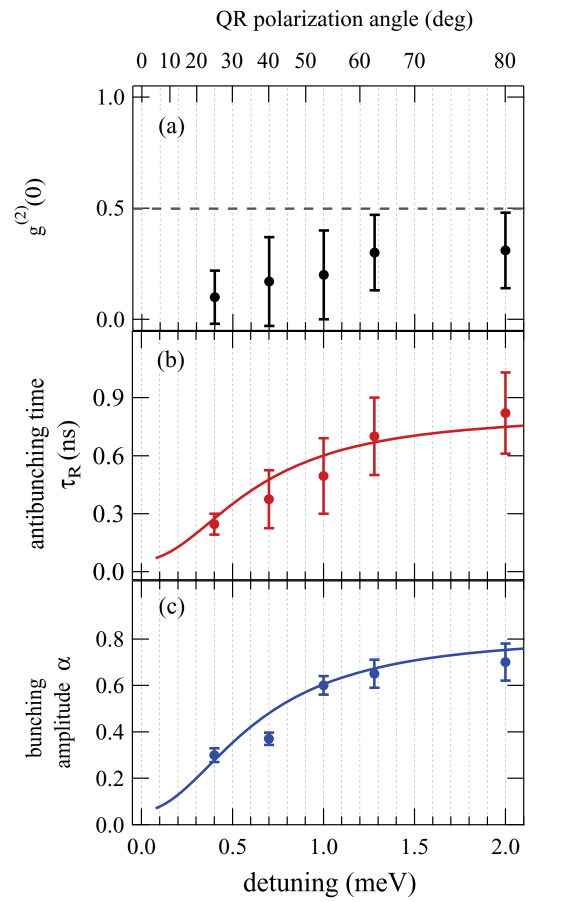

FIG. 3. (Color online) Extracted values of (a) $g^2(0)$, (b) antibunching time τ_R, and (c) bunching amplitude α vs detuning from the results of Fig. 2. The black dashed line in (a) indicates the SPE limit, while the solid lines in (b) and (c) are fits with Eqs. (5) and (6), respectively.

including the Purcell effect [(1)]:

$$\frac{1}{\tau_R}(\delta) = G + \gamma(\delta), \quad (5)$$

where the pump rate G is 0.62 ns^{-1}, resulting from the fit of Fig. 3(b). In this way, we can successfully describe the τ_R behavior vs detuning at constant G. The τ_R decrease for decreasing detuning due to the Purcell effect is in addition to the more studied case of increasing G, either by increasing the excitation power or by excitation at higher energy states of the QD.[29]

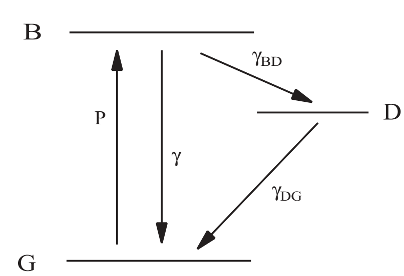

FIG. 4. Schematic energy scheme showing the ground (G), bright (B), and dark (D) exciton states, as well as the transition rates between them.

Bunching describes the enhanced probability of emitting a photon right after a previous one. It has been observed in molecules[26,30] and diamond centers[9,31] as well as in semiconductor QDs.[32] It is generally attributed to part-time occupation of the exciton in a metastable, nonradiative state. In our case, the natural candidate to metastable state is the exciton dark state, although other possibilities as charged excitons cannot be excluded even if they are not observed in the PL spectra. Bunching has been also reported in InP QDs associated with increased absorption rate under excitation resonant with the QD p states.[29] A similar increase can in principle be produced also by refilling the dot from the cavity. This mechanism is compatible with the metastable state model, as it would mainly affect the effective pumping rate G. Spectral diffusion has been recently reported[33] as a bunching mechanism in CdSe QDs. In our case, we do not believe this mechanism to be crucial to our results, as the present PL width (about 150 μeV) is one order of magnitude smaller than the one reported for CdSe QDs[34] and therefore indicates a much weaker spectral diffusion. Bunching due to the increase of the excitation power, as observed in InAs QDs,[35] is also excluded in our experiment, as the power was kept constant. The bunching amplitude α from (2) is shown in Fig. 3(c). It shows a monotonous increase as the detuning increases. The expression for the bunching amplitude is:[26]

$$\alpha(\delta) = \frac{G \cdot \gamma_{BD}}{\gamma_{DG} \cdot (G + \gamma(\delta))}, \quad (6)$$

where γ_{BD} is the bright-to-dark exciton transition rate and γ_{DG} is the dark-to-ground-state exciton nonradiative transition rate. The pumping power is kept constant, and the rates to and from the dark state are not detuning dependent, so the only dependence of $\alpha(\delta)$ on detuning is through $\gamma(\delta)$. The corresponding plot [blue line in Fig. 3(c)] is in good agreement with the experimental data, showing that the interplay between bright and dark excitons together with the Purcell effect is likely to be responsible for the observed behavior. From the fit, we obtain $\gamma_{\mathrm{BD}}/\gamma_{\mathrm{DG}} = 1.62$, which is consistent with a long-lived dark state although a charged exciton could also be responsible for this behavior.

IV. SUMMARY

In summary, we present PL and photon correlation measurements for an InAs QD weakly coupled to a photonic crystal microcavity. The coupled QD is a single photon emitter, whose energy detuning from the cavity simultaneously controls the linear polarization angle of the emitted light and the photon statistics, namely the $g^2(0)$ value, the antibunching time, and the bunching amplitude.

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