

Photon Antibunching from a Single Quantum-Dot-Microcavity System in the Strong Coupling Regime

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We observe antibunching in the photons emitted from a strongly coupled single quantum dot and pillar microcavity in resonance. When the quantum dot was spectrally detuned from the cavity mode, the cavity emission remained antibunched, and also anticorrelated from the quantum dot emission. Resonant pumping of the selected quantum dot via an excited state enabled these observations by eliminating the background emitters that are usually coupled to the cavity. This device demonstrates an on-demand single-photon source operating in the strong coupling regime, with a Purcell factor of 61 ± 7 and quantum efficiency of 97%.

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Cavity quantum electrodynamics (CQED), addressing the interaction between a quantum emitter and a cavity, has been a central topic in atomic physics for decades [1–4] and has recently come to the forefront of semiconductor physics [5–8]. If the coupling between the single quantum emitter and cavity mode is strong compared to their decay rates, the emitter and cavity coherently exchange energy back and forth leading to Rabi oscillations. This strong coupling (SC) regime is of great interest for a variety of quantum information applications, especially with a solid-state implementation. A SC QD-microcavity system could lead to a nearly ideal single-photon source (SPS) for quantum information processing, with extremely high efficiency and photon indistinguishability [9]. The same technology could be applied as an interface between a spin qubit and single-photon qubit in a quantum network [10].

SC between a single atom and a cavity was first achieved more than a decade ago [4]. An analogous system in the solid-state is the excitonic transition of a semiconductor quantum dot (QD) together with a semiconductor microcavity. Several groups have recently reported SC between a single (In,Ga)As QD and either micropillar [5], photonic crystal [6], or microdisk [7] resonators. SC can also occur between a single cavity mode and a collection of degenerate emitters, such as an ensemble of atoms or a quantum well [11]. However, in the latter case the behavior is classical: adding or removing one emitter or one photon from the system has little effect.

In previous studies of the QD-cavity SC [5–7] it was argued that the spectral density of QDs was sufficiently low that it is unlikely that several degenerate emitters contributed to the anticrossing. However, it was not verified that the system had one and only one emitter. There was a surprisingly large amount of emission from the cavity mode when the QD was far detuned. It was unclear whether this emission originated from the particular single QD or from many background emitters. An important step to

establish SC in solid-state CQED is verification that the double-peaked spectrum originates from a single quantum emitter, not a collection of emitters, interacting with the cavity mode.

In this Letter we present proof that the emission from a strongly coupled QD-microcavity system is dominated by a single quantum emitter. Photons emitted from the coupled QD-microcavity system at resonance showed a high degree of antibunching. Away from resonance, emission from the QD and cavity modes was anticorrelated, and the individual emission lines were antibunched. The key to these observations was to resonantly pump the selected QD via an excited QD state to prevent background emitters from being excited. These background emitters, which are usually excited by an above-band pump, prevent the observation of antibunching by emitting photons directly into the cavity mode and by repeatedly exciting the QD after a single laser pulse. With pulsed resonant excitation, the device demonstrates the first solid-state single-photon source operating in the strong coupling regime. The Purcell factor exceeds 60 and implies very high quantum efficiency, making such a device interesting for quantum information applications.

Planar cavities were grown with Bragg mirrors consisting of 26 and 30 pairs of AlAs/GaAs layers above and below a GaAs cavity. A layer of InGaAs QDs, with an indium content of about 40% and a density of 10^{10} cm^{-2} , was grown in the central antinode of the cavity. The QDs typically show splittings between the *s*-shell and *p*-shell transition energies of 25–30 meV, suggesting lateral QD dimensions of 20–30 nm [12]. The cavities were etched into circular micropillars with diameters varying from 1 to 4 μm . An electron microscope image of a 1.2 μm diameter micropillar is shown in Fig. 1(a). Further details on fabrication can be found in Ref. [13].

The coupled-oscillator model gives the complex eigenenergies of the system's two normal modes:

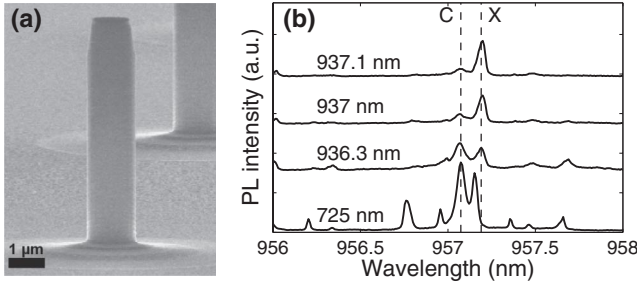


FIG. 1. (a) Scanning electron micrograph of a $1.2 \mu\text{m}$ diameter pillar cavity. (b) Above-band pumping compared to resonant pumping of a chosen QD in Pillar 1. With above-band pumping (725 nm, $0.4 \mu\text{W}$), the chosen QD exciton (X) emits, but so do the cavity (C) and many other QDs. With 937.1 nm ($3 \mu\text{W}$) pump, the chosen QD is selectively excited and its PL dominates an otherwise nearly flat spectrum.

$$E_{1,2} = \frac{E_c + E_x}{2} - i \frac{\gamma_c + \gamma_x}{4} \pm \sqrt{g^2 - \frac{(\gamma_c - \gamma_x - 2i\Delta)^2}{16}}, \quad (1)$$

where E_x and E_c are the energies of the QD exciton and cavity modes, γ_x and γ_c are their full-width half-maxima (FWHM), $\Delta = E_x - E_c$ is the detuning, and g is the exciton-cavity coupling strength. SC requires $g^2 > (\gamma_c - \gamma_x)^2/16$, which leads to a splitting of the two eigen-energies at resonance ($\Delta = 0$) by an amount called the vacuum Rabi splitting. For typical QDs and semiconductor microcavities, γ_x (a few μeV) is much smaller than γ_c ($\sim 100 \mu\text{eV}$), and the SC condition reduces to $g > \gamma_c/4$. In order to reach SC with a given oscillator strength one must maximize the ratio of cavity quality factor to mode volume, Q/\sqrt{V} [5,6]. Our sample showed the highest Q/\sqrt{V} ratio for $1.8 \mu\text{m}$ diameter pillars, which typically exhibited $Q \sim 10000\text{--}20000$, with a mode volume $V_m \sim 0.43 \mu\text{m}^3$.

Photoluminescence (PL) measurements were performed while the sample was cooled to cryogenic temperatures. Increasing the sample temperature caused the QD excitons to redshift faster than the cavity mode, allowing the QDs to be tuned by nearly 1.5 nm relative to the cavity between 6 and 40 K. The sample was optically pumped by a tunable continuous wave (CW) or mode-locked pulse Ti:sapphire laser, focused to a $2 \mu\text{m}$ spot through a 0.75 NA objective. PL was detected by a 750 mm grating spectrometer with N_2 -cooled CCD (spectral resolution 0.03 nm). For photon correlation measurements the PL was spectrally filtered by a 0.2 nm resolution monochromator before entering a Hanbury Brown–Twiss setup [14]. Lifetime measurements were performed using a streak camera with temporal resolution of 25 ps.

In the simplest picture, above-band pumping creates electron-hole pairs that can radiatively recombine to emit photons at the QDs' quantized energy levels. The cavity should be nearly dark if no QD level is resonant with it. However, in previous studies of QD SC the cavity emission

was much brighter than the QD emission even when no QD was resonant with the cavity [5–7]. It was unclear whether the cavity emission resulted from coupling to the specific QD involved in SC, or to a broad background of emitters such as spectrally far-detuned QDs and wetting layer states. These background emitters might contribute to the cavity emission by simultaneously emitting a cavity photon and one or more phonons.

In order to eliminate any background emitters, the laser can be tuned to resonantly pump the excited state (p -shell) exciton in a selected QD [14]. The exciton quickly thermalizes to the QD ground state (s -shell), where it can interact with the cavity. Ideally, resonant pumping creates excitons only in the selected QD, eliminating all extraneous emitters coupled to the cavity.

The PL spectrum of a typical weak coupling device called Pillar 1, excited by CW above-band pumping, is shown in the lowest trace in Fig. 1(b). The cavity mode ($Q = 17300$) could be identified amongst the various QD lines by its broader linewidth, slower tuning with respect to temperature, and lack of saturation at high pump powers. The cavity emits strongly even though there is no QD resonant. The higher traces in Fig. 1(b) show how tuning the pump laser towards an excited state in a chosen QD (937.1 nm in this case) can selectively excite the QD with greatly reduced background cavity emission. Resonant pumping suppresses the cavity emission relative to the QD emission by roughly a factor of 10 in this particular pillar. The resonant pump was nearly 10 times as intense as the above-band pump to achieve the same PL intensity, which caused local heating and led to a slight redshift (0.01–0.03 nm) of the QD line.

The temperature dependent PL for a device exhibiting SC called Pillar 2 is presented in Fig. 2. A clear anticrossing of the QD line and the cavity mode at resonance is evident.

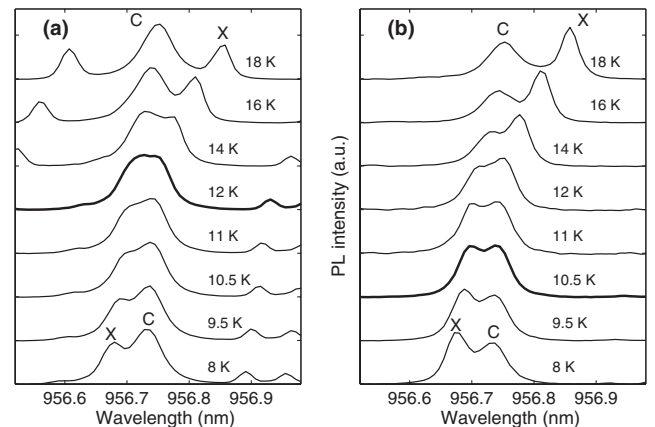


FIG. 2. Temperature dependent PL from Pillar 2 with (a) - above-band CW pump (725 nm), and (b) resonant CW pump (936.25–936.45 nm). Each spectrum is rescaled to a constant maximum since tuning the QD changes excitation efficiency. Resonance occurred at lower temperature for resonant pump case (10.5 vs 12 K) due to local heating.

When the device was pumped above-band (725 nm), the cavity was significantly brighter than the QD and many QDs lines were visible. Resonant pumping of the particular QD involved in SC eliminated the other QD lines and reduced the cavity background emission. The vacuum Rabi splitting at resonance is more pronounced with resonant pumping, possibly because the above-band pump creates background excitons and trapped charges that interact with the QD exciton to broaden its emission.

The line centers and linewidths of the resonantly pumped QD-cavity system [Fig. 2(b)] are shown in Fig. 3. For the lowest temperatures the lower line is narrower and excitonlike, and the upper line is broader and cavitylike. Increasing the temperature causes the lines to switch character as they anticross. From Fig. 3 we determine the cavity linewidth of Pillar 2 is $\gamma_c = 85 \mu\text{eV}$ ($Q = 15200$) and the vacuum Rabi splitting is $56 \mu\text{eV}$. Using formula (1) we calculate $g = 35 \mu\text{eV}$. This gives a ratio of $g/\gamma_c = 0.41 > \frac{1}{4}$ as required to satisfy the strong coupling condition. Fits to the above-band pumped spectra yield a similar value for γ_c , and slightly smaller values for the vacuum Rabi splitting ($50 \mu\text{eV}$) and g ($33 \mu\text{eV}$).

To verify the quantum nature of the system and determine whether a single emitter is responsible for the photon emission, we measured the photon autocorrelation function $g^{(2)}(\tau) = \langle I(t)I(t+\tau) \rangle / \langle I(t) \rangle^2$ of the PL from Pillar 2. With weak excitation, the width of the dip in $g^{(2)}(\tau)$ near $\tau = 0$ is given by the lifetime of the emitter, which is roughly 15 ps (i.e., twice the cavity lifetime) for the resonantly coupled QD-cavity system. The emitter's extremely fast decay rate necessitates a pulsed excitation scheme since conventional photon counters cannot resolve such a short time scale.

The autocorrelation function of photons collected from the coupled QD-cavity system at resonance is shown in Fig. 4(a). The observed value of $g_{r,r}^{(2)}(0) = 0.18 < \frac{1}{2}$ proves that the emission from the coupled QD cavity is dominated by the single QD emitter. Increasing pump power yielded higher values for $g^{(2)}(0)$ as the QD saturated but the cavity emission continued to rise.

Next the QD was red detuned by 0.4 nm from the cavity mode so that photon statistics could be collected from

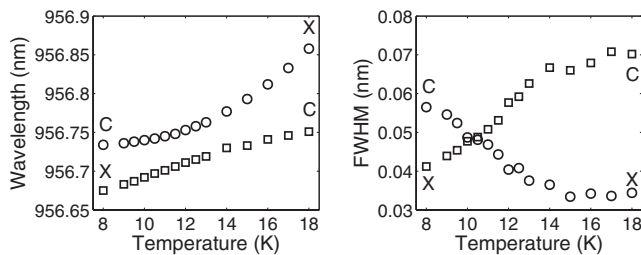


FIG. 3. Emission wavelength and FWHM of upper (circles) and lower (squares) lines as a function of temperature, based on double-Lorentzian fits to resonantly excited spectra of Pillar 2 [Fig. 2(b)].

the cavity and QD emission lines separately. Surprisingly, even with the resonant pump tuned to selectively excite the chosen QD, the cavity emission was ~ 3.5 times brighter than the QD [see Fig. 4(e)]. (Note that with above-band pumping, background emitters were excited and the cavity emission grew another 5 times brighter relative to the QD). The QD emission was antibunched as expected with $g_{x,x}^{(2)}(0) = 0.19$ [Fig. 4(b)]. Interestingly, the cavity emission was also antibunched with $g_{c,c}^{(2)}(0) = 0.39 < \frac{1}{2}$ [Fig. 4(c)], showing that the cavity emission is dominated by a single quantum emitter. This slightly higher value of $g^{(2)}(0)$ suggests that some background emitters were still weakly excited and contribute to the cavity emission. Finally, the cross-correlation function between the QD exciton and cavity emission $g_{x,c}^{(2)}(\tau)$ was measured [Fig. 4(d)]. Strong antibunching was observed with $g_{x,c}^{(2)}(0) = 0.22$, conclusively proving that the single QD emitter is responsible for both peaks in the PL spectrum.

The bright cavity emission cannot be explained by radiative coupling to the QD due to their large detuning. This suggests that another, unidentified mechanism couples QD excitations into the cavity mode when off-resonance. This coupling could possibly be mediated by the absorption or emission of thermally populated acoustic phonons [15, 16].

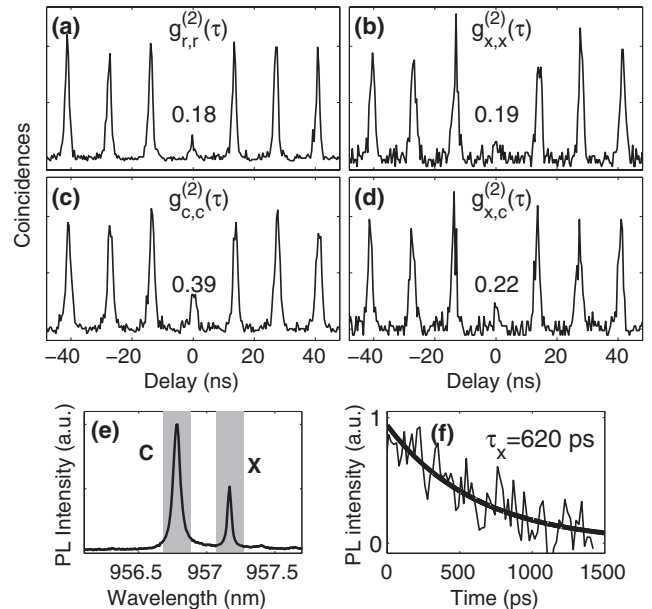


FIG. 4. (a) Measured autocorrelation function of the SC system at resonance, $g_{r,r}^{(2)}(0) = 0.18$. (b)–(e) QD detuned 0.4 nm from cavity. (b) Autocorrelation function of QD emission only, $g_{x,x}^{(2)}(0) = 0.19$. (c) Autocorrelation function of cavity emission only, $g_{c,c}^{(2)}(0) = 0.39$. (d) Cross-correlation function of QD and cavity, $g_{x,c}^{(2)}(0) = 0.22$. (e) PL spectrum. Shaded regions indicate pass bands of spectral filter for correlation measurements. (f) Lifetime measurement of QD only, detuned 0.7 nm from cavity. Dark count backgrounds have been subtracted.

When another SC pillar was pumped with above-band pulses, $g_{r,r}^{(2)}(0)$ of the resonantly coupled QD-cavity system remained between 0.85 and 1 even for the lowest pump powers. Antibunching could not be observed with an above-band pump for two reasons. First, the above-band pump creates many background emitters that couple to the cavity mode, as discussed above. Second, the free excitons created by the pump have lifetimes much longer than the coupled QD-cavity lifetime, allowing multiple capture and emission processes after a single laser pulse. Resonant pumping solves both of these problems.

Under pulsed resonant excitation at the resonance temperature, Pillar 2 emits a pulse train of photons, demonstrating the first solid-state SPS operating in the SC regime. A useful figure of merit for a SPS is the Purcell factor F_P . In the weak coupling limit, F_P gives the enhancement of the QD's emission rate γ due to the cavity: $\gamma = (1 + F_P)\gamma_x$. This relation no longer holds in the SC regime, where the decay rates of the coupled QD-cavity states are fixed at $(\gamma_c + \gamma_x)/2$. We define the Purcell factor more generally as $F_P = \frac{4g^2}{\gamma_c\gamma_x}$ (also called the cooperativity parameter in atomic physics), where γ_x is the QD's emission rate in the limit of large detuning from the cavity. This Purcell factor is often used to quantify the performance of CQED-based quantum information processing schemes [9,17], and is related to the quantum efficiency of the resonantly coupled SPS [18]:

$$\eta = \frac{F_P}{1 + F_P} \frac{\gamma_c}{\gamma_c + \gamma_x}. \quad (2)$$

The efficiency η gives the probability that a photon will be emitted into the cavity mode given that the QD is initially excited. We measured the QD lifetime to be 620 ± 70 ps when the QD was detuned by 0.7 nm from the cavity mode, as shown in Fig. 4(c). At this moderate detuning, the QD's emission rate was slightly enhanced from γ_x by coupling to the cavity. We may calculate the decay rate $\gamma_x = 1/\tau_x$ from formula (1) using $\gamma_{1,2}(\Delta) = 2\text{Im}\{E_{1,2}\}$. From this expression and the measured lifetime, we determine the QD's lifetime in the large detuning limit to be $\tau_x = 700 \pm 80$ ps. This lifetime agrees with measurements of bulk QDs showing an ensemble lifetime of 600 ps when we consider that a pillar microcavity may quench the emission rate of a far-detuned QD by roughly 10% [19]. Using τ_x we determine a Purcell factor of 61 ± 7 and quantum efficiency of $97.3 \pm 0.4\%$.

The high quantum efficiency and short single-photon pulse duration make this device directly applicable to high speed quantum cryptography. However, the incoherent nature of the resonant pump likely results in moderate photon indistinguishability of around 50%. Indistinguishability could be improved using a coherent pump scheme, such as one involving a cavity-assisted spin flip Raman

transition [9,10,17], to make the device ideal for quantum information processing with single photons.

In conclusion, we have observed antibunching in the photon statistics from a strongly coupled QD-microcavity system. The suppressed value of $g^{(2)}(0) = 0.18$ from the system at resonance proves that a single quantum emitter dominates the photon emission. Off-resonance, the QD and cavity emission were both antibunched as well as anticorrelated, further confirming that only one emitter is responsible for the PL. Resonant pumping was essential to these observations, since it eliminated the background emitters that can scatter photons directly into the cavity mode and repeatedly excite the QD after a single laser pulse. Our results demonstrate a solid-state single-photon source operating in the strong coupling regime, with a Purcell factor of 61 ± 7 and quantum efficiency of 97%.

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