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Coherent generation of nonclassical light on chip via detuned photon blockade

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The on-chip generation of non-classical states of light is a key-requirement for future optical quantum hardware. In solid-state cavity quantum electrodynamics, such non-classical light can be generated from self-assembled quantum dots strongly coupled to photonic crystal cavities. Their anharmonic strong light-matter interaction results in large optical nonlinearities at the single photon level, where the admission of a single photon into the cavity may enhance (photon-tunnelling) or diminish (photon-blockade) the probability for a second photon to enter the cavity. Here, we demonstrate that detuning the cavity and QD resonances enables the generation of high-purity non-classical light from strongly coupled systems. For specific detunings we show that not only the purity but also the efficiency of single-photon generation increases significantly, making high-quality single-photon generation by photon-blockade possible with current state-of-the-art samples.

Due to their strong interaction with light and ease of integration into optoelectronic devices, self-assembled quantum dots (QDs) are promising candidates for quantum light sources [1]. High-fidelity single-photon generation from QDs for off-chip applications has been demonstrated under both non-resonant [2] and resonant [3–5] excitation. Some of these experiments have employed micro-pillar cavities [6], etched [7] or epitaxially grown photonic nanowires [8] for enhanced light off-chip extraction efficiency. On the other hand, photonic crystal cavities provide a promising on-chip route toward optoelectronic integration of QDs due to the established set of associated integrated waveguide and detector structures [9, 10]. Such structures will be able to exploit strong light-matter coupling with QDs for the generation of a variety of on-chip non-classical light states by various quantum-electrodynamical (QED) methods, and recent exotic proposals have even explored the possibility of releasing energy exclusively in bundles of n -photons [11]. The phenomena of photon-tunnelling and photon-blockade in strongly coupled systems have been experimentally demonstrated both for the case of the QD on resonance [12–14] and near resonance [15] with the cavity (and likewise, only for resonant atom-cavity system [16]). However, in the case of large detuning these effects have only been investigated theoretically [17].

In this letter, we demonstrate the feasibility of performing photon-blockade at significant detuning, and indeed the importance of doing so for high-purity and high-efficiency operation. We show that by detuning the QD and cavity resonances while operating in the photon-blockade regime, the second-order autocorrelation func-

tion ($g^{(2)}(0)$) of the light transmitted through the cavity decreases from $g^{(2)}(0) = 0.9 \pm 0.05$ to $g^{(2)}(0) = 0.29 \pm 0.04$. Simulations of the second- and third-order autocorrelation functions for our system are in excellent agreement with the measurements, and they reveal that not only does the quality of the single photon stream increase, but that the absolute probability of obtaining a single photon increases by a factor of ~ 2 . Furthermore, we show that the values we obtain for $g^{(2)}(0)$ are only limited by the system parameters (QD-cavity field coupling strength g and cavity field decay rate κ), and that high-quality single-photon emission is within reach for current state-of-the-art samples for specific cavity and QD detunings.

The sample investigated is schematically illustrated in figure 1a and consists of a layer of low density InAs QDs grown by molecular beam epitaxy and embedded in a photonic crystal L3 cavity [18]. The energy structure of a QD strongly coupled to a cavity is well described by the Jaynes-Cummings (JC) Hamiltonian

$$H = \omega_a a^\dagger a + (\omega_a + \Delta) \sigma^\dagger \sigma + g(a^\dagger \sigma + a \sigma^\dagger) \quad (1)$$

where ω_a denotes the frequency of the cavity, a the annihilation operator associated with the cavity mode, σ the lowering operator of the quantum emitter, Δ the detuning between quantum emitter and cavity, and g the emitter-cavity field coupling strength. The resulting eigenenergies, the Jaynes-Cummings-ladder dressed states, are illustrated in figure 1b. For n photons in the cavity the energy is $n\omega_a$ (red lines), and the energy of the quantum emitter (orange) varies with a detuning parameter. Due to the coupling, the resulting energy eigenstates are the anticrossing polariton branches. At resonance, the splitting is given by $2g\sqrt{n}$ (with n being the index of the rung). While this letter explicitly discusses

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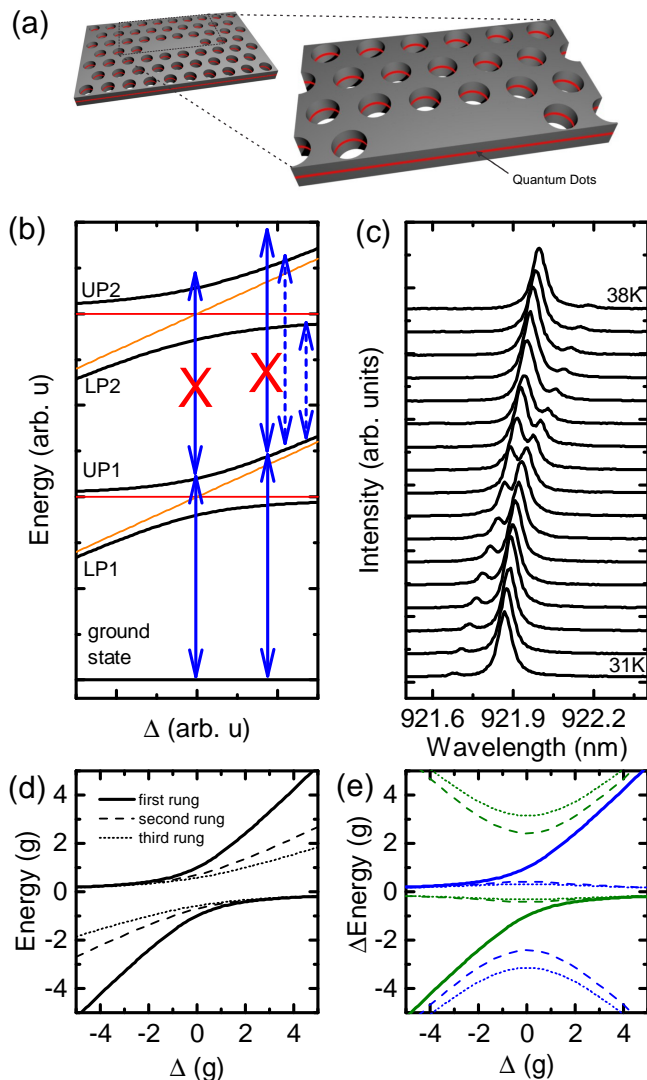


FIG. 1. (a) Schematic illustration of self-assembled QDs embedded in a photonic crystal cavity. (b) Jaynes-Cummings ladder obtained from equation 1 (c) Cross-polarised reflectivity spectrum of the coupled QD-cavity system obtained for tuning the QD through the cavity resonance. (d) Energies for exciting the n -th rung of the Jaynes-Cummings ladder in an n -photon process. (e) Transient energies for climbing the Jaynes-Cummings-ladder rung by rung. Transitions from upper and lower polaritons are colour coded in blue and green, respectively. In panels (d) and (e) the energy of the bare cavity was subtracted from all transitions for better comparison. Δ is the QD-cavity detuning and g the coupling strength.

the case of a QD in a photonic crystal cavity, the same physics holds for a large number of systems such as those formed by atoms [19, 20] or superconducting circuits [21].

For QDs, the anticrossing that results from the coupling to a cavity can be efficiently studied in optical spectroscopy experiments, where the QD and cavity detuning is controlled by the lattice temperature [22, 23]. The result of such a measurement is presented in figure 1c,

which shows the transmission through the cavity measured in a cross-polarised reflectivity configuration [24] in the temperature range $T = 31 - 38$ K. A clear anticrossing provides evidence of strong coherent coupling between QD and cavity. A fit (not shown here) reveals a coupling strength of $g/2\pi = 10.9$ GHz and a cavity field decay rate $\kappa/2\pi = 10.0$ GHz.

Due to the unequal energy spacing (anharmonicity) of the Jaynes-Cummings ladder, transmission of a laser through the cavity affects the beam's photon statistics and introduces strong photon correlations [12, 15]. This is schematically illustrated by the solid blue arrows in figure 1b; if the laser is tuned into resonance with one of the polariton branches of the first rung, it cannot excite the system to the second rung due to the ladder anharmonicity. Therefore, in this regime the transmitted beam consists of a series of single photons and hence is called the photon-blockade regime. However, the fidelity of this process is inherently limited by the transition linewidth, given by the cavity field κ and quantum emitter γ decay rates. In particular, due to final state broadening and the shorter lifetime of excited states, transitions to higher rungs have larger linewidths, further reducing the probability of generating single photons.

Importantly, operating the system at a significant QD-cavity detuning can lead to higher-purity single-photon emission. We consider two cases to support this conclusion: the excitation of a higher rung in a multi-photon process and subsequent excitation. Therefore, we plot the energies for an n -photon excitation of the n -th rung in figure 1d and the transient energies from one rung to the next in figure 1e. Clearly, at zero detuning the energies for exciting the first and higher rungs are close together (figure 1d), and their separation strongly increases for the upper (lower) polariton branch for positive (negative) detunings of the quantum emitter. For a laser in resonance with the first rung the probability of n -photon excitation of higher rungs decreases with increased detuning. Similar scenarios can be found for subsequent climbs up the ladder, as presented in figure 1e, which shows the transition energies from the ground state to the first rung, the first to the second rung and the second to the third rung as solid, dashed and dotted lines, respectively. Transitions from an upper (lower) polariton branch to higher rungs are colour coded in blue (green). Near resonance the first and second transitions are close in energy but their separation strongly increases with the detuning of the quantum emitter (c.f. blue arrows in figure 1b). The close proximity of the first rung to the outer higher order transitions for large detunings does not reduce the single-photon emission character, since these transitions occur from the other polariton branch as can be seen from the different colours. Therefore, a detuning between quantum emitter and cavity is also expected to improve the purity of single-photon generation under photon-blockade for subsequent rung excitation.

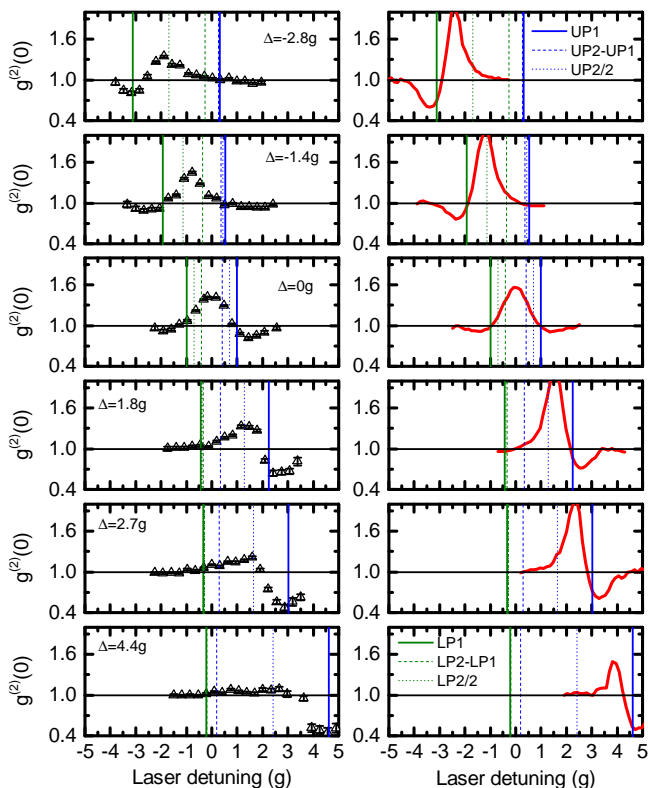


FIG. 2. $g^{(2)}(0)$ as function of the laser detuning for a set of different QD-cavity detunings: (left) experiment and (right) simulation. With increased detuning the depth of the anti-bunching is more pronounced. Vertical lines represent the relevant transition energies of the JC ladder as described in figure 1.

Furthermore, detuning also affects the linewidths of the states in such a way that the linewidth of a polariton branch that evolves towards the bare QD (bare cavity) transition decreases (increases). This further reduces the overlap of transitions involving different rungs of the JC ladder and increases the fidelity of photon-blockade (see supplemental material).

To quantify the quantum character of light the second-order autocorrelation function [25, 26]

$$g^{(2)}(0) = \frac{\langle m(m-1) \rangle}{\langle m \rangle^2} \quad (2)$$

is a commonly used quantity, where m signifies a number of detections in the photocount distribution. It results in a $g^{(2)}(0)$ of 1 for a coherent source and 0 for a perfect stream of single photons. To test our expectation that the purity of single-photon generation under photon-blockade can be improved by detuning the QD and cavity resonances, we measured $g^{(2)}(0)$ from the output correlations of a laser beam transmitted through the cavity. The result of these experiments is presented in the left part of figure 2 that shows $g^{(2)}(0)$ as a function

of the laser detuning for six different QD and cavity detunings. The data were recorded under pulsed excitation with $t_p = 30$ ps long pulses. This pulse duration was chosen as a compromise between frequency resolution and avoiding re-excitation of the system. In the case of $\Delta \approx 0$, the form of $g^{(2)}(0)$ is nearly symmetric with photon tunnelling generating a maximum of $g^{(2)}(0) = 1.45 \pm 0.05$ in the centre, and photon-blockade generating a minimum dip of $g^{(2)}(0) = 0.85 \pm 0.05$ ($g^{(2)}(0) = 0.92 \pm 0.05$) at the laser detuning of $1.5g$ ($-1.5g$). When detuning the QD, the maximum of $g^{(2)}(0)$ shifts such that it stays between the polariton branches before it disappears for detunings greater than $\sim 4g$. The dip of $g^{(2)}(0)$ both moves with and shifts toward the polariton branch that is closer to the bare QD transition. Most strikingly, the depth of the dip increases and reaches a value as low as $g^{(2)}(0) = 0.45 \pm 0.05$ for the detunings of $\Delta = 2.7g$ and $\Delta = 4.4g$. This value is lower than 0.5, indicative of strong single-photon character, and lower than $g^{(2)}(0)$ measured in any prior photon-blockade experiments in the solid state. We note here that since the lifetime of the polariton branch closer to the bare QD transition increases with detuning (for details see supplemental material), excitation with 70 ps long pulses was possible without re-exciting the system at detunings of $\Delta = 3 - 5g$, further reducing antibunching to $g^{(2)}(0) = 0.29 \pm 0.04$ (see supplemental material). Small asymmetries in the experimental measurements result from the wavelength dependence of the cross-polarized laser suppression, asymmetries in the spectral shape of the laser pulse, drift of the QD-cavity detuning, and temperature tuning between curves.

To support our findings, we performed quantum optical simulations using a quantum trajectory method (see supplemental material). The results of these simulations are presented on the right side of figure 2. Overall, the simulations are in excellent qualitative agreement with the measurements and also quantitatively resemble the values measured in the photon-blockade regime. Only small differences exist: the measured maximum values of $g^{(2)}(0)$ are slightly lower than the simulated ones. This can be explained by blinking of the quantum emitter [15], which was not included in the simulations.

To further investigate the single-photon character of the light transmitted through the cavity we performed measurements of the third-order autocorrelation function $g^{(3)}(0) = \frac{\langle m(m-1)(m-2) \rangle}{\langle m \rangle^3}$, as higher-order autocorrelations are necessary to characterise the multi-photon nature of non-classical light [27]. The result of these measurements are presented in figure 3a, which shows $g^{(2)}(0)$ and $g^{(3)}(0)$ as a function of the laser detuning for the case of QD-cavity detuning of $\Delta = 0$ (left) and $\Delta = 2.8g$ (right). Clearly, $g^{(3)}(0)$ shows the same qualitative shape as $g^{(2)}(0)$ but with stronger non-classical values. Simulations of these autocorrelations are presented in figure 3b and show good agreement with the measurements. In

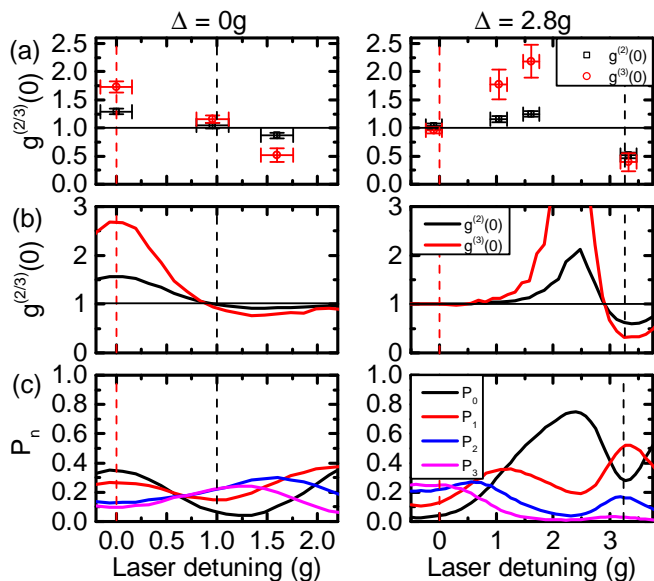


FIG. 3. (a) Measured second- and third-order autocorrelation functions as a function of the laser detuning for a QD detuning of $\Delta_{QD} = 0$ (left) and $\Delta_{QD} = 2.8g$ (right). (b) Simulation for the experimental conditions presented in (a). (c) Simulated probabilities for having $n = 0 - 3$ photons in the output of a pulse transmitted through the cavity. Clearly, for a detuning of $\Delta_{QD} = 2.8g$ (right) P_1 (red) exhibits a pronounced peak.

particular, for the photon-blockade regime the values of $g^{(3)}(0)$ are lower than those of $g^{(2)}(0)$, indicating that $g^{(2)}(0)$ is mainly limited by two-photon events and not higher photon events.

Since the agreement with the measured autocorrelation functions is very good, we can rely on the simulations to explicitly access quantities only within reach of the theory, such as the probabilities P_n of transmitting n photons per excitation pulse through the cavity. These probabilities are presented in figure 3c for $n = 0 - 3$ under the same conditions as the data presented in figure 3a and 3b. Interestingly, we find that in the case of zero QD and cavity detuning (figure 3c left), we see significant contributions of one, two and three-photon events for all laser detunings. In fact, the probability for two-photon events (blue) actually dominates over the probability for single photons (red) in the case of the best photon-blockade. In strong contrast, for a QD-cavity detuning of $\Delta = 2.8g$ (figure 3c right) and operation in the photon-blockade regime, single-photon events (red) strongly dominate over two-photon events (blue) and the probability for three-photon events (purple) becomes negligible. Most strikingly, in the detuned case, not only does the quality of the single-photon stream increase, but the absolute probability of finding a single photon in the transmitted laser pulse increases by a factor of ~ 2 . In addition to the agreement between measured and simulated values for both $g^{(2)}(0)$ and $g^{(3)}(0)$, the experimental

count rates support this finding. Since the overall count rate is proportional to $\sum nP_n$, it does not directly correspond to the overall single-photon efficiency. However, we can still calculate the ratio of the count rates at different detuning conditions in order to compare simulation and experiment. For the points of best photon blockade in the resonant and detuned case, our simulated count rates result in a ratio of $2.27 : 1.05 = 2.16$, which is in very good agreement with the measured ratio of $4 \times 10^4 : 1.8 \times 10^4 = 2.22$.

This counter-intuitive finding that the efficiency of single-photon generation increases when detuning cavity and QD can be understood in the following way: Photon-blockade is obtained if the first rung of the JC ladder is excited while the overlap of the laser with higher rungs is suppressed. When on-resonance this suppression is inherently limited by the linewidth of the transitions, that scales with n as the decay rate of a rung is proportional of the number of photons. Meanwhile, the detuning of subsequent rungs scales with \sqrt{n} (see supplemental material). Therefore, for any system parameters κ and g that can be achieved with the emitter and cavity in resonance, there will always be an overlap between the transition to the first rung and to higher climbs up the ladder. As a result, the strongest photon-blockade with the emitter and cavity in resonance is not observed for the laser exactly on resonance with the first rung of the JC-ladder ($\sim \pm g$), but rather with the laser off-resonant and detuned to $\sim \pm 1.5g$ (c.f. figure 3 - left). In contrast, if the separation between different JC rungs is enhanced by detuning the emitter and cavity, the strongest photon-blockade is obtained with the laser resonant with the polariton branch, making photon-blockade more efficient than in the resonant case. Therefore, not only the purity but also the efficiency of single-photon generation improves given the correct detuning between cavity and emitter. With increasing detuning between the QD and cavity, the oscillator strength of the more QD-like polariton branch decreases as the oscillator strength of the QD is much weaker than the one of the cavity. Therefore, for too large detunings the efficiency decreases, resulting in an *optimum detuning* for single-photon generation of a few g (see supplemental material).

This approach to photon-blockade has strong potential for single-photon generation under already achievable system parameters. Improvements in the spatial alignment of the QD and cavity field have enabled the coupling strength to reach values up to $g/2\pi = 40$ GHz [28]. Recent nanofabrication improvements have allowed for experimental GaAs photonic crystal cavity loss rates as low as $\kappa/2\pi = 4.0$ GHz [29]. When using these parameters in our simulations we obtain $g^{(2)}(0) = 0.1$ in the photon-blockade regime, and an absolute probability of over 90% for single-photon emission, demonstrating that high-quality single-photon streams generated by photon-blockade are within reach.

In summary, we have demonstrated that QD-cavity detuning is a key ingredient for high-purity generation of non-classical light from strongly coupled systems. We have shown that detuning strongly reduces the spectral overlap with higher rungs of the Jaynes-Cummings-ladder and hence greatly improves the generation of single photons by photon-blockade. We have presented quantum-optical simulations that are in excellent agreement with our measurements and show that high-quality single-photon generation under photon-blockade is possible with current state-of-the-art samples. The generation of single photons by photon-blockade might have advantages over other techniques. First, the use of high quality photonic crystal cavities promises a method of on-chip routing of the photons by coupling them to photonic crystal waveguides (with high efficiency) [30]. Second, the cavity emission rate is at least one order of magnitude faster than the bare QD emission rate, resulting in a comparable increase in the maximum single-photon generation rate while maintaining potential advantages from resonant excitation. Furthermore, the successful experimental demonstration of photon-blockade in the detuned light-matter configuration demonstrates the feasibility of operating cavity QED in such an extreme regime and paves the way for a wealth of other quantum light sources, including those generating n -photon states [11].

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