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Photon antibunching from few quantum dots in a cavity

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Single quantum dots (QDs) are frequently used as single-photon sources, taking advantage of the final exciton decay in a cascade that produces energetically detuned photons. We propose and analyze a new concept of single photon source, namely a few-QD microcavity system driven close to, but below the lasing threshold under strong excitation. Surprisingly, even for two or three QDs inside a cavity, antibunching is observed. To quantify the results, we find that a classification of single-photon emission in terms of antibunching in the autocorrelation function $g^{(2)}(0)$ is insufficient and more details of the photon statistics are required. Our investigations are based on a quantum-optical theory that we solve to obtain the density operator for the quantum-mechanical active medium and radiation field.

Single-photon sources are key components in quantum-information technologies. They enable the realization of fundamental quantum-mechanical concepts in current applications [1], such as quantum key distribution (QKD), quantum teleportation, and Bell measurements. To generate single photons, in many practical applications sources based on parametric down conversion are used [2–4]. Alternatively, a single emitter can be driven with a pump pulse to emit exactly one photon on demand per excitation cycle like a turnstile device [5]. Semiconductor quantum dots (QDs) possess distinct properties that make them suited for applications as single-photon sources, such as the possibility of electrical pumping [6]. Resonant excitation schemes have been used to get closer to the ideal case of an isolated "atom-like" single-photon emitter [7]. When using a single QD, a preceding photon from the biexciton recombination can be used to herald the single photon from the exciton recombination, which is always second in the cascade. The extremely high single-photon purity (close to the ideal single-photon Fock-state) obtained from these sources comes at the cost of low repetition rates that are limited by the free-space radiative lifetime of hundreds of ps [1, 8].

QD microcavity laser devices have been demonstrated to exhibit non-classical features in the light emission around threshold [9, 10]. In the following, we explore this quantum-regime for single-photon operation from a single- to few-emitter QD medium. The presence of a cavity enhances the emission rate via the Purcell effect [11], and operation in the GHz regime has been demonstrated for a single QD in a micropillar resonator [12]. The prospect of achieving single-photon emission with several QD emitters coupling to the cavity reduces the requirement on device fabrication to rely on samples with only a single QD.

For single-QD microcavity devices, we identify an optimal balance between cavity-loss rate and light-matter coupling strength that maximizes the emission rate and the degree of antibunching seen in $g^{(2)}(0)$. Next, we explore devices with up to three QDs and quantify their performance in terms of achievable repetition rates, purity of single-photon emission, and antibunching. We see that $g^{(2)}(0)$ fails in uniquely classifying single-photon emission, as the same degree of purity of single-photon emission can be reflected by different values of $g^{(2)}(0)$. Before results are presented, we begin by reviewing the definition and interpretation of $g^{(2)}(0)$ and present the theoretical model.

From the density operator $\rho(t)$ one can obtain the photon statistics for a single mode of the quantized electromagnetic field

$$p_n = \rho^{\mathrm{phot}}_{nn} \,, \tag{1}$$

where $\rho^{\mathrm{phot}}_{nm} = \mathrm{tr}_{\mathrm{el}}\ \rho_{jj'nm}$ is the photonic density matrix obtained by tracing out the electronic degrees of freedom. The photon statistics contains direct information on the single-photon emission probability p_1 and is therefore of central interest. Frequently single-photon performance is expressed in terms of the autocorrelation function $g^{(2)}(0)$ [13–15]. This is for good reason, since measurements of the second-order photon correlation function $g^{(2)}(\tau)$ are well-developed using Hanbury-Brown and Twiss (HBT) setups. The single-mode second-order photon correlation (or autocorrelation) function $g^{(2)}(0) = (\langle n^2\rangle - \langle n\rangle)/\langle n\rangle^2$ is determined by the first two moments ($k = 1$ and 2)

$$\langle n^k\rangle = \sum_{n=0}^{\infty} n^k p_n \tag{2}$$

of the photon statistics. We emphasize that $g^{(2)}(0)$ is therefore an *averaged* quantity that can only be interpreted correctly in regimes where the photon statistics is known (either from other indicators, or from intuition). A well known example is where the single field mode is represented by a Fock state $|n\rangle$. In this case, $g^{(2)}(0) = 1-1/n$, which implies that a single-photon Fock state exhibits $g^{(2)}(0) = 0$, and a two-photon Fock state $g^{(2)}(0) = 0.5$ [16]. For single and few-emitter microcavity systems, especially below and around threshold, the photon statistics is neither Poissonian, nor corresponds to a Fock state, and the probability of realizing a single-photon state in comparison to multi-photon states cannot be directly inferred from the autocorrelation function. In quantum-optics, the identification of non-classical states of light is an active research branch, and various methods have been suggested to obtain access to the probability distribution beyond the grasp of second-order HBT measurements [10, 17–21].

a. Theory. Our theoretical investigations are based on a microscopic semiconductor model that describes the coupled system of few-QD emitters and cavity mode. We solve the von Neumann-Lindblad equation

$$\frac{\partial \rho}{\partial t} = -i[H,\rho] + \sum_X \mathcal{L}_X(\rho) \tag{3}$$

for the density operator ρ of the coupled electronic and photonic degrees of freedom numerically. As a basis, we use the direct product $|j_1,\ldots,j_N,n\rangle$ of the configurations $|j_1,\ldots,j_N\rangle$ of the electronic states of N QDs and the Fock states $|n\rangle$ of the cavity mode. The light-matter interaction is created by the Tavis-Cummings Hamiltonian $H = H_0 + g\sum_{\alpha,\{ij\}}(b^\dagger Q^\alpha_{ij} + \text{h.c.})$, where g is the coupling strength, $b^\dagger$ is the photon creation operator with respect to the cavity mode, and $Q^\alpha_{ij} = |i\rangle_\alpha\langle j|_\alpha$ is a projection operator acting on the configurations i and j of QD α. The summation over $\{ij\}$ encompasses all transitions between configurations that involve a recombination of a QD s-shell electron-hole pair. The free part $H_0 = \sum_{\alpha,i}\varepsilon^\alpha_i Q^\alpha_{ii} + \hbar\omega b^\dagger b$ contains the cofiguration energies in all QDs and the contribution from the electric field. The last term in Eq. (3) gives the Lindblad contributions describing dissipative influences of the environment, which will be discussed later.

The size of the Hilbert space grows rapidly with increasing QD number, but allows for a numerical solution of a small number ($\approx$ 4-5) of emitters fully accounting for correlations between the multi-exciton states of each emitter and amongst emitters, as well as emitter-photon and photon-photon correlations. Correlations between electrons in the QD states and photons determine the statistical properties of the emitted radiation. The feasibility of factorization-based methods, where these correlations are taken into consideration, have been demonstrated for the calculation of threshold-properties of microlasers [9, 22]. Correlations between carriers in different QD emitters are responsible for superradiant coupling effects [23]. Factorization-based approaches rely on a decreasing contribution of correlations between an increasing number of particles in order to facilitate a truncation. In a system with only a few QD emitters, this criterion is not fulfilled. For the present work, we have verified the necessity to use the numerically much more demanding direct solution of the von Neumann-Lindblad equation.

In the following calculations, we use typical material parameters for InGaAs/GaAs QDs, see the caption to Fig. 1. For each QD emitter we account for two confined single-particle states each, for electrons and holes. The ground state configuration is given by the filled valence- and empty conduction-band states. By considering different possibilities to place electron-hole pairs in the single-particle states, one arrives six possible configurations for each QD: two bright states close to the s-shell transition energy, three dark states, and the ground state [24, 25]. All N emitters are connected by the light field (radiative coupling), so that 6^N configurations represent the coupled system and are used in the calculation. We assume the contributing QD transitions to be fully resonant with the cavity mode. Off-resonant coupling and line broadening have been shown to facilitate efficient coupling at high carrier densities and to compensate for slight detunings from the cavity [26–28].

The embedding of self-assembled QD emitters in the surrounding semiconductor material enables capture and relaxation of charge carriers due to Coulomb- and phonon-mediated scattering processes. Each scattering process X is accounted for by a reservoir-interaction Lindblad term

$$\mathcal{L}_X = \frac{\gamma_X}{2}(2X\rho X^\dagger - X^\dagger\rho X - \rho X^\dagger X) \tag{4}$$

in the von Neumann-Lindblad equation (3). Here, γ_X is the associated rate, ρ is the density operator of the coupled many-emitter-photon system, and the operators X act either on the electronic configurations or the cavity photons, depending on the nature of the dissipative process. The Lindblad formalism ensures a consistent treatment of scattering and dephasing, i.e. scattering into and out of a state causes dephasing of coherent polarizations involving this same state [29].

Off-resonant excitation into the wetting layer is modeled as capture of electron-hole pairs into the highest confined QD states at rate γ_P. Scattering from p- to s-shell takes place at rate γ_r, and photons leave the cavity at rate κ. The Lindblad-forms are constructed accordingly [24]. As intra-QD carrier relaxation rates, we use $\gamma_r = 0.1/\text{ps}$. For the situation under study, i.e. strong excitation into saturation at elevated temperatures, this value lies in the range between 3–10 ps confirmed by recent experiments and quantum-kinetic calculations [30, 31].

The limitation to four localized single-particle states per QD serves well for the suggested scenario. It incorporates the main features of multi-level QD systems instead of two-level systems. Since excitation is strong, the system emits from a higher multi-exciton state, whereas the interplay of different multi-exciton states is not so relevant. In this respect, increasing the number of localized states will not add any new physics, but will make a numerical solution unnecessarily challenging, as we solve the full von-Neumann equation for the full state space of a coupled 3-QD (six configurations each)-photon system.

b. Emission from a single QD in a cavity. In Fig. 1 we characterize different operational regimes for a single QD in a microcavity by showing input/output curves and the photon autocorrelation function versus pump rate. Consistent with most experiments, operation in the weak-coupling regime is assumed, i.e. dissipation exceeds the light-matter coupling. The blue curves depict essentially perfect single-photon operation, with $g^{(2)}(0) \approx 0$ for the entire excitation range. This is possible because with a cavity loss rate of $\kappa = 5/\text{ps}$, much faster than the

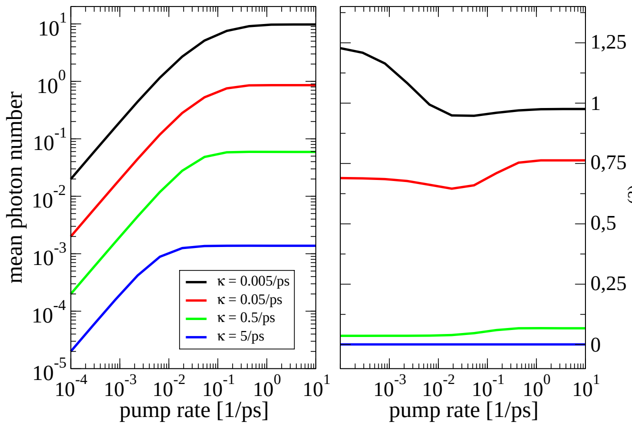

FIG. 1. **Left:** Mean photon number and **Right:** zero-delay second-order photon correlation versus pump rate. Cavity losses are 0.005, 0.05, 0.5, and 5/ps from top to bottom. Other parameters are the light-matter coupling constant $g = 0.1$/ps, and carrier relaxation rate $\gamma_r = 0.1$/ps. The results in all remaining figures are obtained for a pump rate of 10/ps, which is far into the saturated regime.

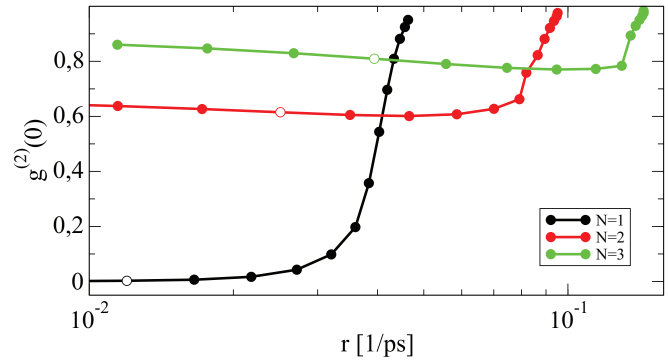

FIG. 2. Autocorrelation function $g^{(2)}(0)$ as a function of the photon output rate for N=1,2,3 emitters. Each curve is obtained from a series of calculations where the cavity-Q is varied. From left to right, κ = 10, 6.3, 4, 2.5, 1.6, 1, 0.63, 0.4, 0.25, 0.16, 0.1, 0.063, 0.04, 0.025, 0.016, 0.01/ps, open circles indicate $\kappa = 2.5$/ps on each curve. High excitation is used to drive the system into saturation.

optical recombination time, there is practically no cavity-storage effect. The down side is a low photon emission rate, as given by

$$r = \kappa \langle n \rangle \ . \tag{5}$$

The second lowest input/output curve ($\kappa = 0.5$/ps) shows improvement in the emission rate from a stronger cavity effect. As long as the cavity lifetime is sufficiently short, no significant photon population can build up in the cavity mode. Thus, photon statistics remains close to that of the cavity-less single emitter. The top two curves in Fig. 1 for $\kappa = 0.05$/ps and 0.005/ps show that while photon number further increases with lower κ, antibunching (quantified by $g^{(2)}(0) < 1$) is severely degraded. The combination of low cavity loss and high excitation can even drive the single emitter into lasing [15, 24], indicated by the black curves approaching $g^{(2)}(0) = 1$ and a mean photon number above unity at high pump rates. It is worth noting that in all cases, saturation of the single QD emitter is reached, above which emission properties remain unchanged even if the pump rate is further increased.

Since strong excitation maximizes the photon emission rate r, we now focus on this regime and compare the attainable degree of antibunching as a function of the attainable emission rate. With the correct combination of cavity-Q and light-matter coupling strength, it is possible to maximize the emission rate far beyond the free-emitter case without cavity, and yet maintain a desired level of antibunching. For the single emitter, represented by the black curve in Fig. 2, results indicate $g^{(2)}(0) < 0.5$ for emission rates up to 40 GHz (0.04/ps), which relates to the pulse-repetition rate for photon-on-demand sources. As an advantage, this result is insensitive to variations in pump rate.

c. Emission from multiple QDs in a cavity. It is widely believed that single-photon purity degrades drastically when there is more than one emitter, to the extent that photon statistics becomes totally classical. This has e.g. been demonstrated by Agarwal et al. when studying resonance fluorescence in atoms [32]. Even in state-of-the-art QD-microcavity samples, background emitters are often present and couple even non-resonantly to a cavity mode [15, 26, 27, 33]. In the following we address the question whether such samples can still constitute efficient single-photon sources. To obtain a quantitative assessment of the problem, we repeat the calculations for two and three identical QDs that are all resonant with the cavity mode. The results are summarized by the upper two curves in Fig. 2. Naturally, the emission rate increases beyond that of the single-QD limit when two or three QDs emit at the same time. It is interesting to see that both cases exhibit non-classical antibunching with $g^{(2)}(0)$ as low as 0.6 in the two-QD case.

In the following, we reexamine the results in terms of *single-photon purity* η (the inverse η^{-1} is the error)

$$\eta = \frac{p_1}{\sum_{i \geq 2} p_i} \ , \tag{6}$$

which we define by separating the probability p_1 to have a single photon from the probability to find multiple photons. Accessing all elements p_n of the photon statistics is possible in the density-matrix formalism. In contrast to $g^{(2)}(0)$, this quantity, together with the emission rate, allow to assess single-photon performance irrespective of the underlying photon statistics. Note that similar parameters have been considered elsewhere, e.g. in [34, 35].

In the left panel of Fig. 3 the purity is compared to $g^{(2)}(0)$ on the horizontal axis. For a microcavity with one, two or three QDs single-photon production with $\eta > 1000$ is possible. However, there is no one-to-one correspondence between η and $g^{(2)}(0)$ for systems of dif-

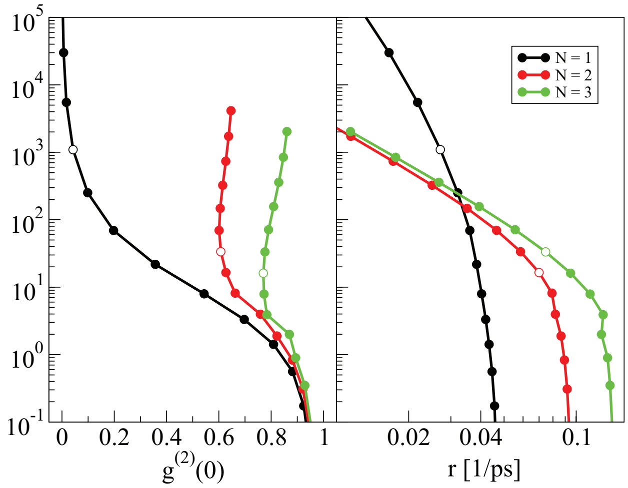

FIG. 3. Single-photon purity η vs. $g^{(2)}(0)$ **(left)** and vs. output rate r **(right)** for N=1,2,3 emitters in a cavity. From high to low η, $\kappa = 10$, 6.3, 4, 2.5, 1.6, 1, 0.63, 0.4, 0.25, 0.16, 0.1, 0.063, 0.04, 0.025, 0.016, 0.01/ps, open circles indicate $\kappa = 0.63$/ps on each curve.

ferent QD numbers. As an example, a three-QD microcavity system exhibiting $g^{(2)}(0) \approx 0.7$ is as good as a single-QD microcavity system exhibiting $g^{(2)}(0) \approx 0.4$ in terms of purity/error. This demonstrates that in a few-emitter microcavity system, where statistical properties of the emission are subject to cavity-QED and few-emitter effects, $g^{(2)}(0)$ fails as measure for the performance of single-photon emission and may, in fact, even give misleading results when used as sole indicator.

Device relevant and central result is the right panel of Fig. 3 showing η versus emission rate r. Comparing the cases for one, two and three emitters, all exhibit the possibility of large η (here $> 10^3$) in the limit of vanishing cavity effects, and achieved at the expense of lower emission rate. Higher purity can only be obtained with the single-QD system ($\eta > 10^5$ in the same limit). For practical applications, such high values of the purity are often not required, especially if the emission rate can be increased at the same time. For example, for a value of $\eta \approx 100$, which can be achieved with one, two or three QDs, the two and three-emitter microcavity systems even offer an enhancement in emission rate up to a factor of two in comparison to the single-emitter system. At the highest output rates, η becomes smaller than unity for any number of emitters, showing that the photon statistics evolves into a Poisson distribution where the peak resides no longer at the origin. Then lasing takes place and puts a boundary on the single-photon operational regime.

d. Conclusion We have investigated the potential of strongly excited few-QD microcavity systems as single photon sources. Strong excitation ensures highest possible repetition rates and insensitivity to pump-rate fluctuations. We map out the tradeoff in device performance in terms of emission rate and single-photon purity for cavities with different Q factors. Surprisingly, single-photon purity up to 1000 (1‰ of the emitted photons come in bunches of two and more) can be achieved with two and three emitters coupling to the same cavity mode. Purities around 100 and less even benefit from a few-emitter gain medium in terms of emission rate, and we predict an attainable rate of up to $\approx 70\,$GHz. Our results on single-photon purity in the presence of several emitters (single emitter plus few additional "unwanted" emitters) has practical value, as it indicates that the requirement for samples to be free of residual emitters is less strict than typically assumed.

While antibunching is characterized in terms of $g^{(2)}(0)$, it fails to quantify the performance of the proposed few-QD microcavity single-photon source. There is no one-to-one correspondence between single-photon purity and $g^{(2)}(0)$ for different number of QDs in the cavity. For the two- and three-QD case, $g^{(2)}(0)$ exhibits non-classical values *above* 0.5, whereas for the same degree of single-photon purity, the single-emitter case satisfies $g^{(2)}(0) < 0.5$. The origin of this failure lies is the fact that in the proposed operational regime, strong cavity-QED and few-emitter effects let the photon statistics deviate from that of a pure Fock, thermal or coherent state.

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