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Nonclassical radiation from a single self-assembled InAs quantum dot

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We report the experimental observation of photon antibunching in fluorescence from a single self-assembled InAs quantum dot. The nonclassical photon statistics proves the atomlike nature of a single quantum dot at cryogenic temperatures. Pump-power-dependent measurements of the correlation function $g^{(2)}(\tau)$ provide a method to determine the recombination time of the single exciton ground-state transition that does not require pulsed lasers. Using this method, we investigate the influence of photonic environment on a single quantum dot exciton lifetime.

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The appearance of sharp lines in photoluminescence has been regarded as the hallmark of full three-dimensional confinement of excitons in various *quantum dot* (QD) structures.[1–3] The underlying atomlike two-level structure of a single QD exciton ground-state transition should at the same time give rise to nonclassical light emission, i.e., photon antibunching and sub-Poissonian photon statistics. After a photon is emitted from a single two-level emitter, the system is necessarily in the radiatively inactive ground state and a second photon cannot be emitted immediately after the first one. The emission probability *recovers* its mean value only after the excited-state occupancy reaches the steady-state population, determined by the excitation and relaxation rates. This dead-time (or waiting-time) between successive photon emission events leads to photon antibunching and sub-Poissonian photon statistics. Since these nonclassical effects are washed out when the number of emitters is larger than unity, the detection of strong antibunching provides a direct evidence that the emission source is a single two-level quantum emitter with an anharmonic energy spectrum.

An experimental method for determining the photon statistics of a quantum emitter is the measurement of the normalized (second-order) intensity correlation function $g^{(2)}(\tau)=\langle :I(t)I(t+\tau):\rangle/\langle I(t)\rangle^2$, where :: denotes normal ordering and $I(t)$ is the intensity operator. So far, photon antibunching has been observed in many different single quantum emitters: a single atom,[4] a single stored ion,[5] and a single molecule.[6] Recently, photon antibunching was demonstrated in solid-state systems: for a single chemically synthesized CdSe quantum dot[7] and a single nitrogen-vacancy center in diamond.[8,9]

In this rapid communication we report the experimental observation of photon antibunching in the fluorescence from the ground-state transition of a single self-assembled InAs QD. The measured values of the correlation function $g^{(2)}(0)$ as low as 0.05 prove that a single self-assembled QD can be considered a strongly anharmonic quantum emitter at cryogenic temperatures. Our results thus show that self-assembled QD's, which possess the advantage that their emission properties can be tailored by their geometrical dimensions and the photonic structures they are placed in, are well suited for quantum optics experiments.

We also show that pump-power-dependent measurements of $g^{(2)}(\tau)$ can be used to determine the fundamental recombination and excitation times of a single QD. As such, these measurements (based on continuous-wave laser excitation) provide an alternative to time-resolved measurements based on mode-locked lasers.[10–12] We use these photon correlation measurements to show that the emission lifetimes of single QD's in an unprocessed sample differ by a large factor (up to ~ 6) from the lifetimes of QD's that are placed in a microdisk cavity but do not couple to any of the microdisk modes. We attribute this lifetime difference to the modification of the density of optical modes available for spontaneous emission and the zero-point fluctuations in the two geometries.[13,14]

Two samples of self-assembled InAs QD's were grown by molecular-beam epitaxy (MBE). Each was based on a AlAs/GaAs short-period superlattice and a GaAs buffer layer for substrate smoothing. The first sample (Sample A) consisted of 100 nm GaAs, an InAs QD layer, and 100 nm GaAs. The second sample (Sample B) was a microdisk structure with a 0.5 μm $Al_{0.65}Ga_{0.35}As$ post. The diameter of the disks was 5 μm and their composition was identical to Sample A. Details of the microdisk structure and the processing can be found elsewhere.[15] The QD's in both samples were grown using the partially covered island technique in order to shift their ground state to higher energies.[16] The emission wavelengths of the QD's were in the range from 925 to 975 nm. The QD's had a diameter of $\sim 400-500$ Å and a height of ~ 30 Å . Both samples exhibited a gradient in the QD density reaching from $\lesssim 10^8$ cm^{-2} to $\sim 10^{10}$ cm^{-2} across the samples.

Our experimental setup consisted of a combination of a low-temperature diffraction-limited scanning optical microscope for spatially resolved photoluminescence spectroscopy and a Hanbury Brown and Twiss (HBT) setup for photon correlation measurements. The QD samples were mounted in a He-flow cryostat and cooled to 4 K. The cryostat was moved by computer-controlled translation stages, thus allowing for scanning across the sample. The QD's were optically excited with a continuous wave TiSa laser operating at 760 nm, generating electron-hole pairs in the GaAs barrier layer which are subsequently captured by the QD's within a short timescale (<35 ps).[17] The same microscope objective (NA

$=0.55$) mounted outside the cryostat was used to both excite the QD's (diffraction limited spot size: ~ 1.7 μm) and collect the emitted fluorescence light. The collected light was spectrally filtered by a 0.5 m monochromator and transferred to the HBT setup, consisting of a 50/50 beamsplitter and two single-photon-counting avalanche photodiodes (APD, EG&G model SPCM-AQR-14, dark count rates <50 s^{-1}). A pinhole was placed in front of each APD to prevent crosstalk between the APD's due to avalanche photons generated by the detection events of the photodiodes. The APD's were connected to the start and stop inputs of a time to amplitude converter (TAC). The TAC output was stored in a multichannel analyzer (MCA) to yield the number of photon pairs $n(\tau)$ with arrival time separation of $\tau = t_{start} - t_{stop}$. An electronic delay of 12 ns was introduced into the stop channel in order to check for the symmetry of the photon correlation around $\tau = 0$. The time resolution of the HBT setup was 420 ps.

Single QD's were identified by scanning across the sample (Sample A) or scanning across the microdisks (Sample B) and checking the emission spectrum for characteristic sharp lines. The pump-power dependence of the emission spectrum was carefully checked in order to exclude the occurrence of emission lines from weakly excited additional QD's. We also verified that the emission intensities decreased when the pump laser spot was moved by an amount consistent with the diffraction limit. The total detection efficiency for the radiation emitted from a single QD was estimated to be 5×10^{-5}, including the extraction efficiency of the radiation from the material, the collection efficiency of the microscope objective, the transmission of optics and monochromator, and the quantum efficiency of the APD's.

The measured photon count distribution $n(\tau)$ was normalized to the expectation value for counting completely uncorrelated photons obeying a Poissonian arrival time distribution. The normalized measured distribution $\overline{n}(\tau)$ is equivalent to the correlation function $g^{(2)}(\tau)$ as long as the measured time separation τ between photon pairs is much smaller than the mean time ΔT_D between detection events.[18] As $\tau \leqslant 10^{-7}$ s and $\Delta T_D \geqslant 3\times10^{-5}$ s in all experiments, this condition was easily fulfilled.

Figures 1(a) and 1(b) shows the measured photon count distribution $n(\tau)$ for the 1X transition of a single QD on sample A for two different pump intensities. These intensities correspond to an excitation of the QD well below saturation (a) and at the onset of saturation (b). Saturation is defined here as the pump intensity at which the 1X line reaches its maximum intensity.[19] Traces (a) and (b) exhibit a clear dip in the correlation counts for a time delay $\tau = 0$, indicating a strong photon antibunching. The values of the normalized count distribution $\overline{n}(0)$ are 0.32 and 0.46 for traces (a) and (b), respectively. These values are limited by the time resolution of our photon counting setup. In order to correct for the time resolution we fitted the normalized count distribution $\overline{n}(\tau)$ with the correlation function $g^{(2)}(\tau) = 1 - ae^{-|\tau|/t_m}$ convolved with a Gaussian time distribution with 420 ps full width half maximum. The parameter a accounts for the background present in the measurements. The resulting fitted $g^{(2)}(\tau)$ is shown as solid line in Fig. 1. The values of $1-a = g^{(2)}(0)$ obtained from the fit are 0.23 and 0.34 for traces (a) and (b). The fact that $g^{(2)}(0) < 0.5$ in both traces unambiguously indicates that the measured photon antibunching from the 1X transition stems from a single, anharmonic quantum emitter.

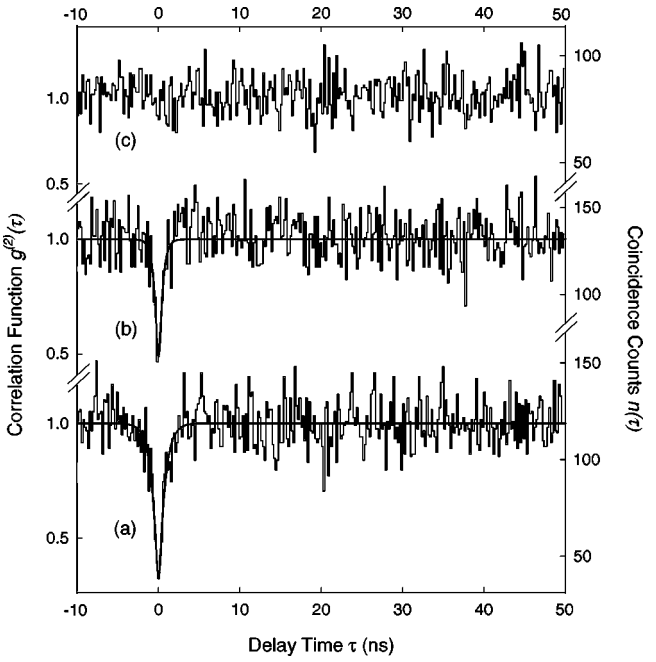

FIG. 1. Measured distribution of coincidence counts $n(\tau)$ and fit of the correlation function $g^{(2)}(\tau)$ (solid line) for a single QD of Sample A, obtained at two different pump intensities: 66 W cm^{-2} [Trace (a)] and 252 W cm^{-2} [Trace (b)]. Trace (c) shows the coincidence counts for many QD's in a high density region of Sample A. The time bin is 195 ps, the integration times and single APD count rates are: $T_a = 3600$ s, $N_a = 13\,000$ s^{-1}; $T_b = 3154$ s, $N_b = 14\,500$ s^{-1}; $T_c = 1761$ s, $N_c = 15\,000$ s^{-1} for traces (a), (b), and (c), respectively.

The measured correlation function $g^{(2)}(0)$ does not reach its theoretical minimum of zero because of the presence of background straylight. The correlation function $g_b^{(2)}(\tau)$ expected in the presence of a background radiation is $g_b^{(2)}(\tau) = 1 + \rho^2(g^{(2)}(\tau) - 1)$ where $\rho = S/(S+B)$ is the ratio of signal S to background B counts.[9] From optical emission spectra of the single QD we can determine ρ for the 1X transition for the two pump intensities to be 0.9 and 0.83, respectively. The resulting values of $g_b^{(2)}(0)$ are 0.18 and 0.30, which are in good agreement with $g^{(2)}(0)$ values determined by the fits. The antibunching time constants t_m obtained from the fit of $g^{(2)}(\tau)$ are 736 ps and 444 ps for traces (a) and (b). For comparison, trace (c) of Fig. 1 shows the correlation for multiple emission lines of many QD's in a high-density region of sample A. This correlation is flat over the complete measurement time and its normalized value of 1 corresponds to a Poissonian photon statistics. Here, many independent (nonidentical) QD's contribute to the measured correlation and signatures of single QD antibunching are completely washed out.

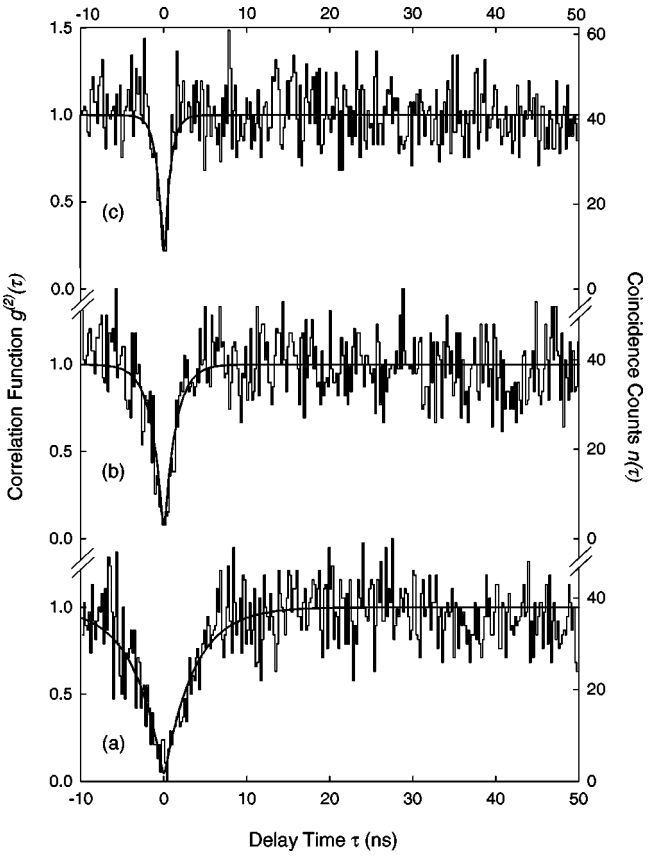

FIG. 2. Measured distribution of coincidence counts $n(\tau)$ and fit of the correlation function $g^{(2)}(\tau)$ (solid line) for a single QD located in a microdisk cavity of Sample B, obtained at three different pump intensities: 17 W cm^{-2} [Trace (a)], 56 W cm^{-2} [Trace (b)], and 105 W cm^{-2} [Trace (c)]. The time bin is 195 ps, the integration times and single APD count rates are: $T_a=5615$ s, $N_a=6000$ s^{-1}; $T_b=3925$ s, $N_b=7000$ s^{-1}; $T_c=4334$ s, $N_c=7000$ s^{-1} for traces (a), (b), and (c), respectively.

Figure 2 shows the measured photon count distribution $n(\tau)$ for the 1X transition of a single QD located in a microdisk cavity of Sample B for three different pump intensities. The pump intensities correspond to an excitation of the QD well below saturation (a), at the onset of saturation (b), and well within saturation (c). We carefully verified that the QD did not couple to a mode of the microdisk resonator: the QD 1X transition exhibits a resolution limited linewidth of 70 μeV whereas the nearest cavity mode (with linewidth 110 μeV) is approximately 3 meV apart. As the background radiation was weak for the QD located in the microdisk, the correlation measurements of Fig. 2 show very strong antibunching. The values of the normalized count distribution $\overline{n}(0)$ are 0.05, 0.08, and 0.22 for traces (a), (b), and (c), respectively. The fit of $g^{(2)}(\tau)$, performed as discussed above, yields values of $1-a=g^{(2)}(0)$ of 0.02, 0.00, and 0.09. The signal to background ratio for the three different measurements was determined to be $\rho=0.98$, 0.98, and 0.94 respectively. The values of $g_b^{(2)}(0)$ expected in the presence of background thus are 0.04, 0.04, and 0.12; these values once again agree very well with the fitted values of $g^{(2)}(0)$. In summary, these data correspond to a perfect antibunching in the radiation from a single QD. The time constants for the exponential decay of the antibunching were determined from the fit to be 3.60 ns, 1.41 ns, and 750 ps for the three pump intensities. A reference measurement for a single QD in an unprocessed part of Sample B also shows strong antibunching with time constants of 870 ps and 812 ps for pump intensities of 60 and 80 W cm^{-2}, corresponding to an excitation below saturation and at the onset of saturation.

From the pump-intensity-dependent time constants measured in the experiments described above we can determine the lifetime of a single exciton ground-state transition. We theoretically model the QD's with a simple three-level rate equation model, based on the model in Ref. 19, taking into account exciton and biexciton transitions and neglecting higher multiexciton contributions as well as nonradiative decay channels, e.g., Auger recombination. Using the quantum regression theorem we find that the time dependence of the normalized correlation function $g^{(2)}(\tau)$ in the low excitation regime is described by a simple exponential decay as given above. From this model we derive that the dead time between photon emission events is given by the excited state population recovery time. Thus, the model explains the strong dependence of the measured lifetimes on the pump intensities. By comparing the predictions of the model with the measured lifetime data we can estimate the lifetime for the single exciton transition $t_{1X}=1/\Gamma_{1X}$ in the limit of a vanishing pump rate to be 900 ± 100 ps (Sample A), 1.10 ± 0.05 ns (Sample B, reference QD in unprocessed part), and 6.0 ± 0.5 ns (Sample B, QD in microdisk). The uncertainties of $\pm10\%$ in the lifetime values are mainly due to fluctuations of the pump intensity during the experiment (slight shifts of the translation stage positions cause a change of the effective pump rate) and the uncertainty in relating the measured pump intensities to pump rates in the theoretical model.

Even though the estimated lifetimes for the bare QD's are comparable to values in the literature,[10] the exciton lifetime of QD's located in a microdisk cavity is considerably longer than the values previously reported. We performed measurements on various microdisks of sample B and found exciton lifetimes in the range from 2.2 to 6.0 ns. In order to independently verify these lifetimes we measured the exciton decay rate by the technique of time correlated single photon counting[20] by pumping the QD's with a modelocked ($\sim$250 fs) TiSa laser at 755 nm and detecting start pulses from a part of the pump beam on one APD and stop events from the QD emission on the other APD. These time-resolved studies gave an exciton lifetime that agrees within a margin of 10% with the values obtained from the antibunching measurements.

The difference in lifetimes could be explained by the different photonic environments in which QD's are located: In Sample A, the thin GaAs film containing the quantum dot layer faces air on one side and the substrate on the other side, whereas for Sample B, the GaAs film is surrounded by air on both sides. For the microdisk QD (Sample B) this leads to a modification of the density of optical modes available for spontaneous emission and zero-point fluctuations of the electrical field.[13,14] From Ref. 13 we find that $t_{1X}^B/t_{1X}^A \propto (n_{sub}^2+1)/2$, where n_{sub} is the refractive index of the substrate and

t_{1X}^{A} and t_{1X}^{B} are the recombination times for QD's located in Samples A (bare QD) and B (QD in microdisk), respectively. This formula is valid for a film thickness L larger than $\lambda/2$, where λ is the material wavelength of the emitted light ($L = 200$ nm and $\lambda/2 = 134$ nm in our case), but L still optically thin with respect to absorption, and is obtained by averaging over emission angles, polarizations, positions, wavelengths, and thickness fluctuations.[13] The validity of the above formula is also confirmed by Ref. 14. For GaAs at 950 nm ($n_{sub} = 3.54$), we obtain a lifetime ratio of 6.8 which gives an upper limit to the ratios of 2.0 to 6.7 obtained from the measurements. The different experimental ratios might be caused by different radial positions of the QD's within in the microdisk resonators (close to post vs close to circumference). However, due to the limited spatial resolution of our setup we cannot precisely determine the QD position relative to the microdisk. Our findings for the enhanced exciton lifetime are further supported by a theoretical calculation in Ref. 21: the spontaneous emission rate from a dipole located in a thin dielectric layer into free modes is strongly suppressed (by a factor of ~ 10 for the parameters of our structures) compared to a dipole embedded in a uniform index material. Emission into guided modes of the thin film can be neglected in our case.[14] We remark, however, that this explanation is tentative since we cannot exclude additional effects that could influence the emission lifetimes of QD's placed in microdisk structures. A thorough theoretical analysis of the emission lifetimes would have to account for the special geometries of the microdisk resonators, e.g., by performing a finite-difference time-domain analysis. Nevertheless, our results indicate that the recombination times of self-assembled QD's are determined largely by radiative recombination because only this process is affected by the different photonic environments.

In summary, we demonstrated that the photoluminescence from a single InAs QD exhibits one of the key signatures of nonclassical light, i.e., an almost perfect photon antibunching. The nonclassical statistics truly proves the atomlike nature of a single QD. From the measured antibunching time constants we could determine the lifetime of the single exciton transition of a single bare QD and a single QD placed in a microdisk structure. The large difference in recombination times could be explained by the different density of optical modes and the different zero-point fluctuations of the electrical field in the two photonic environments. The observation of nonclassical radiation from a single InAs QD constitutes an essential first step in the realization of single photon sources, cavity-QED experiments, and quantum information processing in semiconductor nanostructures.

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