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Physica E 13 (2002) 412–417
PHYSICA E
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# A quantum dot single-photon source

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**Abstract**

We demonstrate a deterministic source for single-photon pulses with nearly 100% efficiency based on pulsed laser excitation of a single-self assembled InAs quantum dot (QD). The single-photon emission at the single exciton ground state transition is ensured by the anharmonicity of the multi-exciton spectrum in combination with slow relaxation of highly excited QD states. We observe single-photon emission both from a free QD and a QD in resonance with a high-quality factor mode of a semiconductor microcavity. In the second scheme, photons are primarily emitted into the cavity mode due to the Purcell effect, which in principle allows for an efficient use of the generated single-photons due to increased output coupling.


*PACS:* 42.50.Dv; 78.67.Hc; 78.55.Cr

*Keywords:* Single-photon source; Photon statistics; Quantum dots; Quantum optics

## 1. Introduction

Generating single-photons "on demand", i.e., realizing a light source that, upon a trigger event, emits *one and only one* photon within a short time interval, resembles the ultimate control of the light emission process. Many applications in the emerging field of quantum information science [1] require such a deterministic source of single-photons, also termed single-photon turnstile device [2]: Single-photon pulses are an indispensable key element for quantum cryptography, allowing for an unconditionally secure distribution of secret keys [1]. As any measurement unavoidably modifies the state of a single quantum system, an eavesdropper cannot gather information of the transmitted key without being noticed. A necessary condition for this scheme is that the pulses used for transmission of the key do not contain two or more photons. Another major goal in the field of quantum information science is the implementation of quantum computing schemes. Here, it has recently been proposed that basic quantum logical gate operations can be performed using single-photons and linear optical elements [3]. The single-photon sources employed today either rely on highly attenuated laser pulses or on parametric down-conversion. In both schemes, however, photons are created randomly and in order to maintain a low two-photon emission probability the

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PII: S1386-9477(02)00156-X

average photon number per pulse has to be kept much below one.

The realization of a single-photon source requires three key elements: a single quantum emitter, control of the emission process and a real world interface. The single quantum emitter needs to possess a high quantum efficiency ($\eta \approx 1$) and has to emit photons one by one with a certain temporal separation. The latter characteristic, known as photon antibunching, has been demonstrated for a large number of single quantum emitters, e.g., a single atom [4], a single stored ion [5] and a single molecule [6]. Recently, photon antibunching was demonstrated in solid-state systems: for single nitrogen-vacancy centers in diamond [7,8], a single chemically synthesized CdSe quantum dot [9] and for self-assembled InAs QDs [10]. Photon antibunching is a necessary but not sufficient condition for a single-photon turnstile device as photons are emitted at random times and not deterministically. Therefore a control of the emission process is required, consisting of an excitation of the single quantum emitter with probability one and subsequent (in most of the cases) spontaneous emission of a photon. The trigger event for emission could be, e.g., a pulsed excitation of the emitter. Spontaneous emission of a quantum emitter in general is directed into the full solid angle and thus hard to capture efficiently. Coupling of the emitter to a cavity mode allows for a directional emission and thus for using the generated photons in external applications due to the enhanced detection efficiency.

A single-photon turnstile device based on a mesoscopic double barrier p–n heterojunction was proposed in 1994 [2] and demonstrated recently [11], where single as well as multiple photon emission events with a repetition rate of 10 MHz at 50 mK have been reported. This scheme utilizes Coulomb blockade of tunneling for electrons and holes in a mesoscopic p–n diode structure to regulate the photon generation process. The device can only be operated at ultra-low temperatures ($T \leqslant 1$ K) as single electron and hole charging energies must be large compared to the thermal background energy to ensure single-photon emission. Triggered single-photon sources based on single molecules have been demonstrated [12,13] whereby regulation of the photon emission process is achieved by combining either adiabatic passage techniques or non-resonant pumping with pulsed optical excitation. More recently, triggered single-photons have been generated by pulsed optical excitation of single quantum dots [14,15]. The photon correlation measurements reported in Refs. [12,13,15] show a significant probability (12%–26%) for two-photon emission due to background radiation.

The single-photon source that we report [14] is based on a single QD embedded in a high-quality factor ($Q$) microcavity structure. The distinguishing feature of our QD single-photon source is the absence of pulses that contain more than one photon. To ensure single-photon generation at the fundamental QD exciton transition (1X), we adjust the pump power so that two or more electron–hole pairs are captured by the QD during each excitation pulse. The capture time of carriers into the QD ($\approx$ 35 ps [16]) is much shorter than the exciton radiative lifetime. The energy of the photons emitted during relaxation depends significantly on the number of multi-excitons that exist in the QD, due to Coulomb interactions enhanced by strong carrier confinement [17]. If the total recombination time of the multi-exciton QD state is longer than the recombination time of the free electron–hole pairs, each excitation pulse can lead to at most one photon emission event at the 1X-transition. Therefore, regulation of photon emission process can be achieved due to a combination of Coulomb interactions creating an anharmonic multi-exciton spectrum and slow relaxation of highly-excited QDs leading to vanishing re-excitation probability following the photon emission event at the 1X-transition [17]. If the QD exciton recombination is predominantly radiative, every excitation pulse from the mode-locked laser will generate an ideal single-photon pulse. In addition, we achieve turnstile operation of the QD emission resonantly coupled to a high-$Q$ microdisk cavity mode. Here the Purcell effect significantly reduces the photon emission time jitter and ensures that photons are primarily emitted into the cavity mode.

## 2. Experimental setup

The microcavity samples were grown by molecular beam epitaxy and consisted of a 5 μm diameter GaAs disk, containing self-assembled InAs QDs (density $\leqslant 10^8\ \mathrm{cm}^{-2}$), and a pedestal area (microdisk structure) [18]. The QDs had a diameter of ≈40–50 nm and a height of ≈3 nm, emitting in the wavelength range from 920 to 975 nm. The cavity modes of the microdisk resonator are whispering gallery modes (WGM) with quality factors ($Q$) of up to ≈ 6500.

The experimental setup combines a diffraction-limited scanning optical microscope for spatially resolved photoluminescence (PL) spectroscopy and a Hanbury Brown and Twiss (HBT) setup for photon correlation measurements. The system provides spectral resolution of 70 μeV, spatial resolution of 1.7 μm, and temporal resolution of 420 ps. The microdisks are mounted in a He gas flow cryostat. Optical pumping is performed with a mode-locked femtosecond (∼ 250 fs) Ti:sapphire laser, operating at 750 nm and generating electron–hole pairs in the GaAs layers. A microscope objective (numerical aperture NA = 0.55) is used to focus the excitation laser onto the sample and to collect the emitted PL from the QDs. The collected light was spectrally filtered by a 0.5 m monochromator. In all our experiments we used the monochromator to spectrally select the QD single exciton (1X) ground state transition and to perform photon correlation measurements on its emission. The HBT setup consisted of a 50/50 beamsplitter and two single-photon-counting avalanche photodiodes (APD, EG& G model SPCM-AQR-14, dark count rates $< 50\ \mathrm{s}^{-1}$). The APDs 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 $\tau = t_{\mathrm{start}} - t_{\mathrm{stop}}$. An electronic delay of 12 ns was introduced into the stop channel. The measured photon count distribution $n(\tau)$ is equivalent to the unnormalized second-order intensity correlation function $G^{(2)}(\tau)$ as long as the measured time separation $\tau$ between photon pairs is much smaller than the mean time $\Delta T_{\mathrm{D}}$ between detection events, which was the case for all experiments. The total detection efficiency for the radiation emitted from a single QD (off-resonance to any microcavity mode) was estimated to be $5 \times 10^{-5}$, 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 APDs.

## 3. Photon correlation measurements

For identification of the QD 1X transition and the microdisk WGM we first recorded PL spectra from the microdisks containing the QDs. Fig. 1 shows such a PL spectrum from a single QD taken under excitation from a continuous wave (cw) Ti:sapphire laser. The resolution limited (70 μeV) line at 1.3222 eV, labeled 1X, appears first at low excitation powers (1 W/cm$^2$) and is identified as single exciton ground state transition. The line at 1.3196 eV shows a superlinear increase with excitation power and originates from a biexciton recombination (2X). The broad (200 μeV) line at 1.3208 eV corresponds to background emission coupled into a microdisk WGM. The inset of Fig. 1 shows the measured normalized cw photon correlation function $g^{(2)}(\tau)$ for the 1X transition of the single QD in the microdisk at the onset of saturation. The dip at $\tau = 0$ arises from photon antibunching [10] and the fact that $g^{(2)}(\tau) < 0.5$ proves that the emitted light stems from a single quantum emitter.

Turnstile operation of the QD emission is now achieved under pulsed excitation: Fig. 2 shows the measured unnormalized correlation function $G^{(2)}(\tau)$ for (A) the pulsed Ti:sapphire excitation laser, and (B) the 1X transition of a QD that is far detuned from all modes of the microdisk resonator

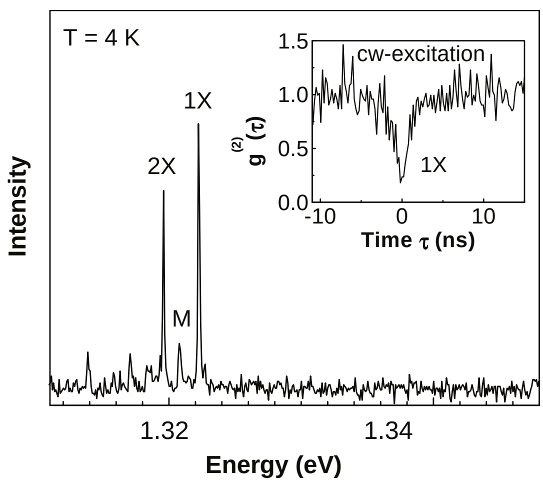


Fig. 1. Photoluminescence spectrum from a single InAs QD embedded in a 5 μm diameter microdisk. Contributions from the single exciton ground state transition (1X), biexciton (2X), and a whispering gallery mode (M) are visible. Inset: Measured cw photon correlation function $g^{(2)}(\tau)$ for the 1X transition.

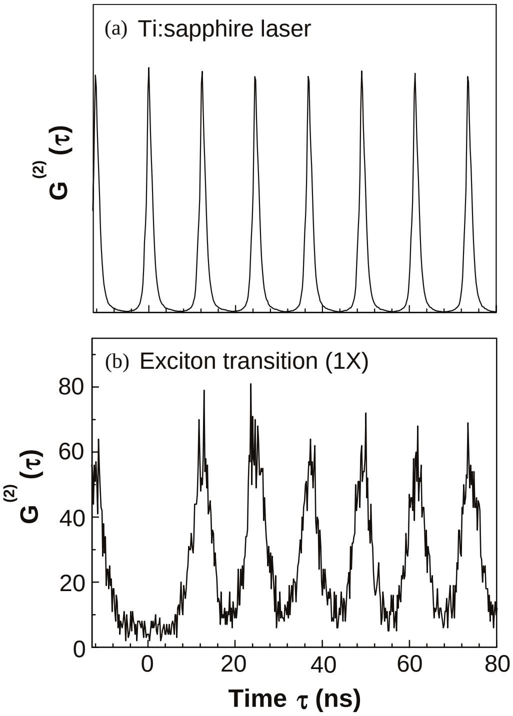


Fig. 2. Measured unnormalized correlation function $G^{(2)}(\tau)$ of (A) a mode-locked Ti:sapphire laser (FWHM = 250 fs), and (B) a single QD excitonic ground state (1X) emission under pulsed excitation conditions (82 MHz).

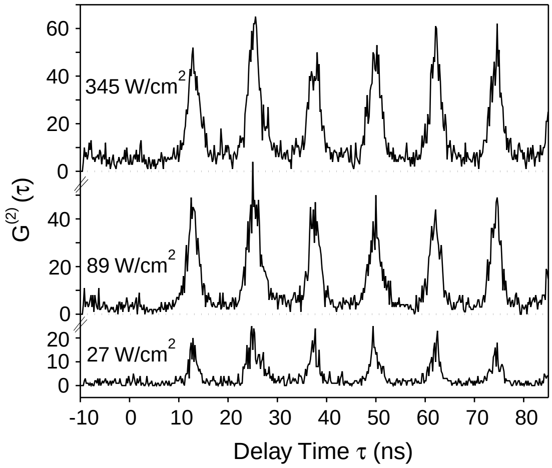


Fig. 3. Measured unnormalized correlation function $G^{(2)}(\tau)$ of a single QD excitonic ground state emission under different excitation powers: (from bottom to top) 27, 89, and 345 $W/cm^2$, corresponding to pump powers below, at, and above the saturation pump power of the single exciton recombination, respectively.

(at temperature $T=4$ K). $G^{(2)}(\tau)$ of the pulsed Ti:sapphire laser exhibits peaks at integer multiples of the pulse repetition rate $T_{rep}$, as one would expect for a pulsed coherent source. The photon correlation of the QD 1X emission also shows peaks at multiples of $T_{rep}$, indicating the locking of the emission to the trigger pump pulses. However, the peak at $\tau = 0$ is no longer present, i.e., within the duration of the pump pulse the probability of finding a second photon following the detection of the first photon at $\tau = 0$ vanishes. Absence of the peak at $\tau = 0$ provides strong evidence for an ideal single-photon turnstile operation. As the QD 1X transition is driven in saturation (pump power where the 1X line reaches maximum intensity) in this experiment the probability of having no injected electron–hole pair in the QD is negligible. Recent experiments show evidence that the recombination is predominantly radiative [10], thus every excitation pulse generates a single-photon pulse. Carriers in the GaAs barrier and the wetting layer recombine much faster than the QD exciton (100–200 ps vs. 2.2 ns [10]) such that the probability of re-excitation of the QD and emission of a second photon is very small ($< 5 \times 10^{-3}$).

Fig. 3 shows the turnstile operation from a single QD for different excitation powers, below, at and above saturation of the single exciton line, respectively. For all pump powers, no peak is visible at $\tau=0$, proving that the turnstile operation is very robust against changes in operation conditions. These results indicate that the probability of two-photon emission per pulse is negligible even when the QD 1X transition is well below or above saturation.

In addition, we achieve turnstile operation of a coupled QD-cavity system by temperature tuning the 1X transition of another QD into resonance with a microdisk whispering gallery mode ($Q\sim6500$) [19]. Fig. 4 shows the measured $G^{(2)}(\tau)$ for the 1X transition (A) out of resonance ($T = 4$ K), and (B) on resonance with the microdisk mode which is reached at a temperature $T = 36$ K. Out of resonance the photon correlation of the QD emission again shows clear evidence for single-photon generation. The correlation peaks are broader compared to Fig. 2B due to the longer 1X recombination lifetime (3.4 ns) of the QD used here. On resonance the FWHM of the photon correlation peaks is narrower by a factor 3.4

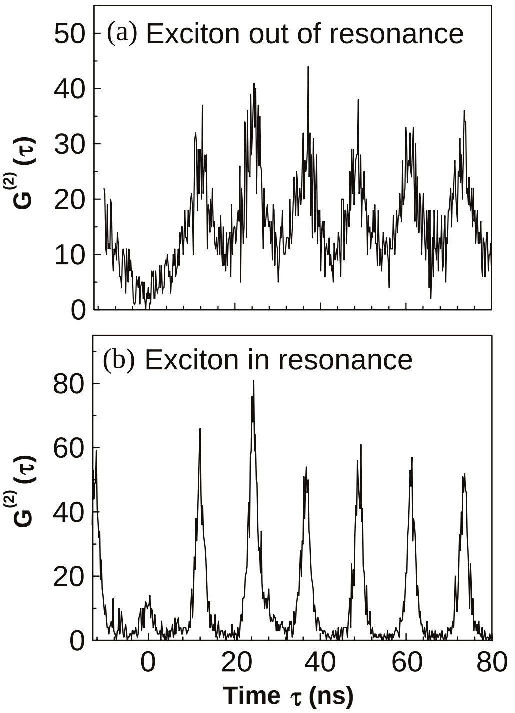


Fig. 4. Measured unnormalized correlation function $G^{(2)}(\tau)$ of a single QD excitonic ground state emission (A) out of resonance, and (B) at resonance with a cavity mode ($Q\sim6500$), under pulsed excitation conditions (82 MHz).

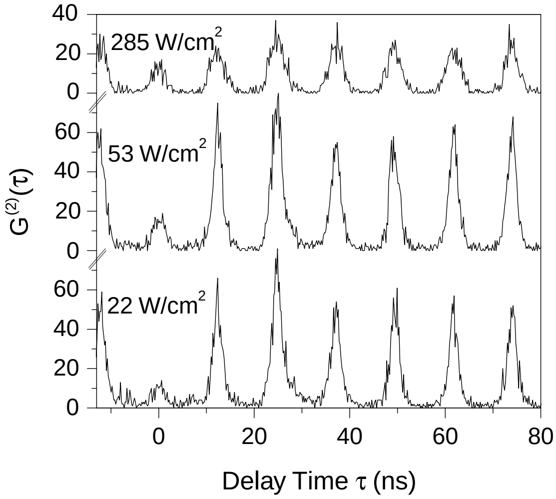


Fig. 5. Measured unnormalized correlation function $G^{(2)}(\tau)$ of a single QD excitonic ground state emission at resonance with a cavity mode, under different excitation powers: (from bottom to top) 22, 53, and 285 $W/cm^2$.

due to the Purcell effect which causes a reduction of the 1X transition lifetime and ensures that photons are primarily emitted into the cavity mode. A small peak at $\tau = 0$ is observed in the resonance case, giving rise to a two-photon emission probability of 29% (given by the ratio of the peak at zero delay time to the peaks at $\tau \neq 0$). This peak could arise due to the Purcell effect, which shortens the 1X recombination lifetime and thus increases the probability of capturing a second electron–hole pair after the first 1X recombination has occurred. In this case multiple photon emission from the 1X transition would happen within the duration of one pump pulse. However, we assume that the main contribution to the zero delay time peak stems from background light generated by the wetting layer or by excited states of other QDs and coupled into the cavity mode. Two experimental observations support this assumption: first, when the QD 1X transition is off resonance the mode emission is still visible in the PL spectrum. Second, we have performed pump power dependent photon correlation measurements of the 1X emission in resonance, displayed in Fig. 5. Here the pump power increases from bottom to top traces, the bottom trace being identical to Fig. 4B. With increasing pump power the relative area of the peak at $\tau = 0$ increases from 0.29 at 22 $W/cm^2$ to 0.56 at 285 $W/cm^2$. This increase of the zero delay time peak is in sharp contrast to the photon correlation pump power dependence of the QD emission far detuned from any cavity resonance (Fig. 3). If the peak at $\tau = 0$ was due to multiple photon emission from the single exciton recombination its relative area should not vary with pump power.

## 4. Summary

In conclusion, we have demonstrated deterministic single-photon emission from a self-assembled single InAs QD placed in a semiconductor microdisk cavity. For a QD far detuned from any cavity resonance, nearly 100% of the excitation pulses lead to the emission of a single-photon with a repetition rate of 82 MHz. If the excitonic recombination is coupled to a high-$Q$ ($Q \approx 6500$) whispering gallery mode up to 70% of the excitation pulses give rise to single-photon emission. Due to the Purcell effect, the time jitter of photon emission is reduced by a factor 3.4, thus theoretically allowing for repetition rates of up to 1 GHz. In principle the Purcell effect could also ensure a higher collection efficiency, provided that the cavity structure allows for a directional output [20]. We envision that the operating temperature of our single-photon source can be easily extended to $T=77$ K enabling use in practical applications. Room temperature operation could in principle be achieved by using QDs with higher confinement potentials to suppress non-radiative carrier losses into the barriers.

## Acknowledgements

We would like to thank B. Gayral for helpful discussions. This work was financially supported by David and Lucile Packard Fellowship, and a grant from ARO. We acknowledge support from the Deutsche Forschungsgemeinschaft (C.B.) and the Max Kade Foundation (P.M.).

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