Single-photon and photon-pair emission from CdSe/Zn(S,Se) quantum dots

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We report on the generation of triggered single photons and photon pairs relying on pulsed optical excitation of epitaxially grown self-assembled CdSe quantum dots (QDs). Single-photon emission is studied on individual CdSe/Zn(S,Se) QDs for temperatures up to 200 K. At low temperatures nearly perfect single-photon emission is achieved whereas at higher temperatures an increasing multi-photon emission probability due to spectrally overlapping acoustic phonon sidebands of neighboring QDs is observed. The multi-photon emission probability of a bare QD (background subtracted) is strongly suppressed. Furthermore, the polarization cross correlation of the biexciton–exciton cascade has been investigated in a CdSe/ZnSe QD at low temperatures. A strong polarization correlation of the emitted photon pairs of ~74% is observed in a linear detection basis.

The rapidly evolving field of quantum information technology requires the development of light sources delivering single photons and pairs of photons on demand. Triggered emission of single photons [1, 2] and photon pairs [3, 4] have already been realized for (In,Ga)As quantum dots (QDs) at low temperatures (T < 50 K). For practical applications operation at high temperatures (T > 77 K), preferentially at room temperature, is important. We report on the generation of triggered single photons obtained from self-assembled CdSe/Zn(S,Se) QDs at temperatures up to 200 K. Furthermore, the polarization correlation of photon pairs generated by biexciton–exciton cascades in a CdSe/ZnSe QD are analyzed at low temperatures.

Two types of CdSe QDs have been grown by migration enhanced epitaxy on GaAs substrates (for details, see [5, 6]). The first type is a CdSe dot embedded in a ZnSe barrier. These QDs emit in the blue spectral region (458–470 nm) and possess a shallow electronic confinement energy (~120 meV). The second type of QDs possesses a Zn(S,Se) barrier and emits in the green spectral range (505–525 nm). These QDs exhibit a higher quantum efficiency at elevated temperatures due to the better electronic confinement of the carriers (~500 meV). In order to select a single QD, small mesas with different diameters down to ~100 nm were fabricated by electron beam lithography and wet chemical etching [7]. The samples were mounted in a temperature variable He-flow cryostat (4–300 K). Our experimental setup combines a diffraction-limited scanning optical microscope for spatially resolved photoluminescence.
cence (PL) and a Hanbury Brown and Twiss (HBT) setup for photon correlation measurements. Both types of QDs were excited by a frequency-doubled mode-locked Ti:sapphire laser (82 or 76 MHz), generating electron–hole pairs in the barrier layers of the QDs. In the case of the CdSe/Zn(S,Se) QDs the light was passed through a monochromator to spectrally filter the excitonic transition of a single QD. In contrast, the light from the CdSe/ZnSe QDs was directly passed to the HBT setup for cross correlation measurements of photon pairs emitted through the biexciton–exciton cascade. Spectral and polarization selection of the biexcitonic and excitonic PL of a single QD was achieved by use of tunable narrow bandpass filters (FWHM = 0.25 nm) and λ/2-polarizers positioned inside the two optical pathways of the HBT setup, respectively. The photons are detected by single-photon counting avalanche photodiodes (SPAD). The electrical pulses of the two SPADs were used to start and stop a time-to-amplitude converter whose output is stored in a multichannel analyser. The measured distribution \( n(\tau) \) is equivalent to the unnormalized second-order correlation function \( G^{(2)}(\tau) \) if the measured time separation \( \tau \) between photon pairs is much smaller than the mean time between detection events, which was always the case for our measurements. Time-resolved PL spectroscopy on single QDs was performed using the Ti:sapphire laser system in combination with a fast photomultiplier (Hamamatsu).

In Fig. 1a the second-order correlation function of the exciton emission line for two different QDs under pulsed excitation (82 MHz). The temperature for the first QD was set to (a) \( T = 4.5 \) K and for the second QD to (c) \( T = 200 \) K. (b) and (d) \( \mu \)-PL spectra of the selected emission line. Vertical bars correspond to the spectral window used for the correlation measurements.

The excitation powers used for these measurements are above the saturation powers of the corresponding exciton emission. For \( T = 4.5 \) K the normalized peak area at \( \tau = 0 \) ns is just 0.04 of those at other delay times. This implies a decrease of multi-photon emission pulses by a factor of 25 in comparison to a Poissonian light source. With increasing temperature the peak area at \( \tau = 0 \) ns increases and reaches a value of 0.81 at 200 K. This increase can be assigned to the stronger dominance of the background (see Fig. 1d) caused by acoustic-phonon sidebands of neighboring QDs [8]. The background

![Fig. 1](online colour at: www.interscience.wiley.com)

Decay characteristics of the exciton (X) (solid circles) and biexciton (XX) (open circles) transition. The solid lines represent model calculations as discussed in the text. Inset: Corresponding \( \mu \)-PL spectrum where the X and XX transition lines of a single CdSe/ZnSe QD dominate. \( T = 4.5 \) K.

![Fig. 2](online colour at: www.interscience.wiley.com)
causes uncorrelated photons which superimpose the correlated photons of the observed excitonic PL. In order to extract the single-photon emission of the individual QD under consideration, the contribution of the background has to be subtracted. For a single-photon source in combination with a Poissonian background the zero delay peak is increased by \(1 - \rho^2\) with \(\rho = S/(S+B)\), where \(S\) is the signal and \(B\) the background [9]. \(\rho\) was determined to be 0.54 ± 0.08 for \(T = 200\) K. The resulting value for \(G^2(\tau = 0)\) is 0.71 ± 0.1 which is in good agreement with the measured \(G^2(\tau = 0)\) value of 0.81 (see Fig. 1c). Therefore, after substraction of the background light the \(G^2(\tau = 0)\) value is 0.1, indicating a suppression of the multi-photon emission probability by a factor of 10 for the individual, bare QD up to 200 K.

While the CdSe/Zn(S,Se) QDs are promising candidates for room-temperature single-photon emission due to the large electronic confinement of the carriers, the CdSe/ZnSe QDs are more suitable for the production of photon pairs, because the biexciton emission in these QDs is typically more pronounced. The inset in Fig. 2 shows a µ-PL spectrum of a single CdSe/ZnSe QD where the exciton (X) and biexciton (XX) transitions are centered at 2.6902 and 2.6739 eV, respectively.

Both transitions reveal a monoexponential decay as can be seen in Fig. 2. Each solid line corresponds to a two-exponential fit using one rise and one decay time, corrected by convolution with the measured time resolution of the detection system (≈140 ps). The estimated decay time of \(t_x = 250 \pm 10\) ps is only slightly larger than the biexciton decay time of \(t_{xx} = 180 \pm 10\) ps. This is a consequence of the spin structure of the recombining particles and the spatial separation of the holes in a QD of finite extension, as shown recently in detail [10].

We further analysed the polarization correlation properties of the photon pairs emitted through the radiative biexciton-exciton cascade of a single QD. The corresponding cross-correlation function of the X and XX emission line is shown in Fig. 3 for four special polarization combinations. We chose a linear detection basis of the polarizers, where H denotes for horizontal and V for vertical detection relative to the lab’s perpendicular. The normalized peak areas at \(\tau = 0\) ns for collinear alignment of the polarizers (VV, HH) of 1.96 (a) and 1.95 (d) reflect a strong bunching behavior while those at perpendicular detection (HV, VH) with peak areas of 0.73 (b) and 0.61 (c) reflect an antibunching effect. This corresponds directly to a pronounced polarization correlation of the emitted photon pairs in a linear basis [3, 4].

A correlation degree of 74.5% can be estimated from the ratio of the peak areas at \(\tau = 0\) under collinear/perpendicular polarization [12]. In addition, it has been found that a tilt of the detection basis of about ±10° leads to a decrease of the correlation degree in both cases. This is consistent with the expected behavior of a linearly polarized cascade emission, where the optical path selection becomes random if the detection angle approaches 45°. The observed strong linear polarization of the photon pairs is a precondition to realize a photon source for passive quantum key encoding [4].

We found no clear indication for an asymmetry of the zero delay time peak shapes as is known for the cascade emission in InAs QDs [4, 11]. This is simply due to the fact that the temporal shape of the correlation peaks is determined by the excitonic and biexcitonic lifetimes [11] which are much shorter in our
case (see Fig. 2) than the time resolution of our HBT setup (~700 ps). However, just this fast dynamical behavior makes CdSe QDs very attractive for triggered single photon and/or photon pair generation at high repetition rates above 1 GHz.

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References