Correlated Photon-Pair Emission from a Charged Single Quantum Dot

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The optical creation and recombination of charged bieexcit and trion complexes in an (In,Ga)As/GaAs quantum dot is investigated by micro-photoluminescence spectroscopy. Photon cross-correlation measurements demonstrate the temporally correlated decay of charged bieexciton and trion states. Our calculations provide strong evidence for radiative decay from the excited trion state which allows for a deeper insight into the spin configurations and their dynamics in these systems.

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In the expanding research fields of quantum information technology the achievement of deterministic, i.e. fully controllable single-photon devices is of highest interest as this provides the key for the realization of novel scalable data processing schemes in quantum computing and cryptography. In recent years, various types of semiconductor-based quantum dot structures have been demonstrated as promising concepts, especially the development of electrically driven sub-Poisson photon sources, which marks an important step.

As was first shown by Moreau et al., single QDs are also capable to serve as sources of temporally correlated photon pairs formed by the cascaded decay of bieexcitonic and excitonic states. More detailed investigations have revealed a strong linear polarization correlation of those photon pairs in consistence with a QD asymmetry-induced lift of the exciton states’ degeneracy.

Besides studies on the creation and decay of neutral (multi-)excitonic carrier configurations in QDs, in recent investigations also the charged counterparts of these complexes have attracted increasing interest due to their possible applications in quantum information processing schemes. In particular, magnetoluminescence measurements have enabled to study the QD trionic fine structure in high detail. Further investigations have also demonstrated coherently manipulated charged QD states.

In this paper we provide direct evidence for the cascaded emission from a charged bie exciton via an excited trion state which is supported by a detailed theoretical analysis. In addition, our studies enable a deeper insight into the fundamental electronic properties of charged QDs and the corresponding optical transitions.

The QD sample under investigation was grown by molecular beam epitaxy (MBE) using n-doped (001)-oriented GaAs as the substrate material. On top of a thin GaAs buffer layer a single layer of self-assembled InAs islands was deposited which formed 3-D confined individual quantum dots (height: 1-2 nm; width: 15-20 nm) within a thin ternary wetting layer (1 ML) after final capping by a 50 nm top layer of GaAs. The QD surface density was $\approx 10^{10}$ cm$^{-2}$. In order to enable studies on individual QDs, an array of single mesa structures was fabricated in a post-growth step by combined electron beam lithography and wet chemical etching. The investigations discussed in the following were performed on a selected 320 nm diameter mesa.

Our experiments have been performed on a combined low-temperature (4 K) micro-photoluminescence (µ-PL) system and a Hanbury Brown and Twiss (HBT)-type photon correlation setup for investigations on the photon emission statistics (for details see Ref. [13]). Spectral selection within our HBT setup was achieved by the use of tunable acousto-optical ($\Delta \lambda = 1$ nm) and/or narrow-band interference filters ($\Delta \lambda = 0.5$ nm) inside the detection paths. The histograms $n(\tau)$ (normalized: $g^{(2)}(\tau)$) of photon correlation events with delay times $\tau$ were measured by a multi-channel-analysers. For quasi-resonant (p-shell) or off-resonant (above GaAs barrier) continuous wave (cw) excitation a Ti:Sapphire laser was used. For pulsed auto-correlation measurements, the laser system was used in its picosecond mode ($\lambda = 2.95$ μm, $\tau = 2$ ps at 76.2 MHz). A steep angle (30°) optical fiber geometry was used for excitation which allowed for focusing down to a spot diameter of ~ 10 μm on the sample surface.

Fig. 1(a) shows µ-PL spectra as obtained for above-barrier cw excitation ($E_{exc} = 1.597$ eV; $P_{exc} = 1.9$ kW/cm$^2$). Apart from an almost unstructured small PL background the spectral window of interest is dominated by a pair of intense narrow lines at 1.3414 eV and 1.3372 eV which exhibit nearly resolution-limited linewidths of 55 ± 10 μeV and 75 ± 10 μeV, respectively. As is depicted in the inset of Fig. 1(a), the PL of the two lines reveals a linear (slope = 0.95 ± 0.05) and super-linear (slope = 2.31 ± 0.05) increase of intensity with excitation density over almost two decades. Together with the observed relation between onset and saturation this reflects the excitonic (X) and biexcitonic (XX) origin of these emission lines. Additional evidence is obtained from time-resolved spectroscopy providing...
a radiative lifetime \( \tau_X \approx 1.0 \) ns and \( \tau_{XX} \approx 500 \) ps for these lines. The assignment of both peaks to the same QD is strongly supported by the observed characteristic line spacing of \( \Delta E_{XX-X} = 4.2 \pm 0.1 \) meV which is consistent with the typical biexciton-exciton Coulomb exchange energy for those dots.

As is shown in Fig. (b), the emission spectrum completely changes when the exciting laser is tuned energetically into the QD p-shell \( (E_{exc}^{res} = 1.4068 \pm 0.0004 \) eV; \( P_{exc} = 1.9 \pm 0.1 \) kW/cm\(^2\)). This quasi-resonant excitation reveals the onset of a new set of almost background-free narrow lines centered at 1.3400 eV \((50 \pm 5 \) µeV), 1.3362 eV \((80 \pm 5 \) µeV) and 1.3342 eV \((110 \pm 5 \) µeV) on the low energy side of the strongly suppressed X and XX PL signals (marked by arrows). In the following, we focus on the two former lines which correspond to the radiative decay of the charged biexciton complex \((1e^22e^1h^2 \rightarrow 1e^12e^1h^1)\) and the consecutive trion recombination from its excited state \((1e^12e^1h^1 \rightarrow 2e^1)\), respectively, as will be demonstrated below.

In order to investigate the photon emission statistics of the observed PL lines we have performed photon auto-correlation measurements using the HBT setup. As an example, the result obtained from the intense XX\(^-\) peak (1.3400 eV) under quasi-resonant pulsed excitation is shown in Fig. (a). Multiple-photon (\( \geq 2 \)) emission is found to be fully suppressed on this PL line which manifests itself in the absence of the zero delay \((\tau = 0 \) ns) coincidence peak. This behaviour demonstrates almost perfect (background-free) triggered single-photon generation from an individual QD.

Furthermore, in Fig (a) the correlation peaks at delay times \( \tau = n T_{\text{laser}} \) \((n = \pm 1, \pm 2, \ldots)\) corresponding to correlation events of photons following subsequent laser excitation cycles \( T_{\text{laser}} = 13.12 \) ns reveal a clear bunching-like behaviour. This means, we observe a higher probability of detecting another XX\(^-\) photon in the next (previous) excitation cycle than in later (earlier) cycles. Such an effect has been reported recently on similar InAs QDs especially under the condition of pure quasi-resonant pumping into an excited state\(^{22}\) and is interpreted as a fast "blinking" process between the neutral and charged state configuration of a single QD. Therefore, the blinking is assumed to reflect the condition of a favored even or odd-number carrier occupancy of the QD. Furthermore, it is important to note that this strongly suggests a residual charging of the QD by ionization of adjacent impurities\(^{23}\) which supports the preliminary assignment of the PL signatures given in Fig. (b). Applying a fit formula\(^{22}\) of the type \( g^2(\tau) = 1 + g_1 \cdot \exp(-\tau/\tau_{\text{blink}})\) for discrete values \( \tau = n T_{\text{laser}} \) \((n = \pm 1, \pm 2, \ldots)\) to the Poisson-normalized correlation peak areas of the data in Fig. (a), we can extract the parameters \( g_1 = 1.67 \pm 0.14 \) for the bunching amplitude and \( \tau_{\text{blink}} = 12.8 \pm 2.0 \) ns for the blinking time constant. The short timescale of this effect is consistent with the generally expected behaviour for a regime of excitation close to the saturation level\(^{22}\).

The assignment of the observed PL under quasi-resonant excitation to charged complexes is supported by a simple model scheme: In the absence of laser excitation a (doping-related or equivalently intrinsic, e.g. donor-type) impurity in the vicinity of an individual QD enables an excess electron to relax into the QD ground state thus initially leaving a charged dot and a nearby ionized impurity. Consequently, under p-shell excitation the creation of odd-number charge states (i.e., e-h pairs plus an extra electron) should be favored. In contrast to this, for above-barrier pumping the effect of photo-depletion\(^{24,25}\) is expected: The dissociation of hot e-h pairs through the local QD-impurity Coulomb field leads to the attraction of holes into the QD whereas excess elec-
trons are also trapped by the impurity centers. In this case, the radiative decay of neutral X and XX complexes is predicted.

In addition, we have performed photon cross-correlation measurements under quasi-resonant continuous-wave (cw) excitation in order to identify the temporal order for the emission of the discussed XX and X− lines. Fig. 2(b) shows the corresponding correlation histogram as measured $n(\tau)$ (left axis) and in the Poisson-normalized $g^{(2)}(\tau)$ (right axis) representation. Using the XX− emission line (1.3400 eV) as the "start" and the X− line (1.3362 eV) as the "stop" trigger of the HBT, a pronounced signal asymmetry was observed in the vicinity of zero delay. The anti-bunching for $\tau < 0$ together with the observed bunching behaviour at positive delays $\tau > 0$ clearly identifies the two decay channels under investigation to be temporally correlated, i.e. to form a radiative cascade originating from the same QD.

For a theoretical analysis of the experimental data discussed above the eigenstates and the emission spectra of the QD have been computed using a full configuration interaction (FCI) procedure. These results indeed show that the XX− emission line appears at a significantly lower energy than the XX− line, while the emission from the trionic ground state (i.e., X−) is placed at slightly higher energies and therefore is not a good candidate.

The charged biexciton PL line is brought below the trionic ground state emission line by the (charged) biexciton binding energy. As a result one finds the following XX− exciton energies $E_{XX}^− = E_A − E_B = E_s^X − \Delta$ for the neutral biexciton and $E_{XX}^− = E_B − E_C = E_s^X − X_{sp}$, with $\Delta$ the (charged) biexciton binding energy. As a result one finds the following values for the emission lines:

$$E_{XX}^− = E_A − E_B = E_s^X − \Delta$$
$$E_{XX}^− = E_B − E_C = E_s^X − X_{sp}.$$
In addition to the experiments discussed before, we have also tested the possibility to generate polarization-correlated photon pairs by the $XX^– \rightarrow X^{– \ast}$ cascade under investigation. Surprisingly, within our experimental accuracy no polarization correlations between the two emitted photons could be observed. Together with the blocked relaxation of the $p$-shell electron in the excited trion configuration (see preceding discussion), these data allow us to get some insight into the spin dynamics in these systems. For higher dimensional systems it is known (see, e.g. Ref. 22) that the fastest relaxation occurs for the hole spin, followed by that of the exciton. It takes the longest time for the electron spin to flip. This hierarchy seems to be maintained also for the QD structures under study: The fact that emission from the charged trion configuration (see preceding discussion), these biexciton carrier complexes have been observed under pure quasi-resonant excitation. Besides the capability of background-free triggered single-photon generation, the cascaded nature of these decay channels was directly demonstrated by means of photon cross-correlation measurements. From a direct comparison with many-particle QD eigenstate calculations, especially for the involved trion complex clear indications for a predominant recombination from its excited state was revealed, thus reflecting a long-lived excess electron spin configuration.

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