Correlated photon-pair emission from a charged single quantum dot

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The optical creation and recombination of charged biexciton and trion complexes in an (In,Ga)As/GaAs quantum dot is investigated by microphotoluminescence spectroscopy. Under the condition of quasiresonant pulsed excitation into the *p* shell we find nearly background-free photoluminescence from single quantum dots where multiple-photon (>1) emission is fully suppressed. This reflects almost perfect triggered single-photon emission. Photon cross-correlation measurements demonstrate the temporally correlated decay of charged biexciton and trion states. Our calculations provide strong evidence for radiative decay from the excited trion state, thus indicating a long-lived *p*-electron spin configuration.

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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.[1] In recent years, various types of semiconductor-based quantum dot (QD) structures have been demonstrated as promising concepts[2–7] where especially the development of electrically driven sub-Poisson photon sources[8,9] marks an important step.

As was first shown by Moreau *et al.*,[10] single QD's are also capable of serving as sources of temporally correlated photon pairs formed by the cascaded decay of biexcitonic and excitonic states. More detailed investigations have revealed a strong linear polarization correlation of those photon pairs[11–13] consistence with a QD asymmetry-induced lift of the exciton states' degeneracy.[14]

Besides studies on the creation and decay of neutral (multi)excitonic carrier configurations in QD's, in recent investigations also the *charged* counterparts of these complexes have attracted increasing interest[15–19] due to their possible applications in quantum information processing schemes. In particular, magnetoluminescence measurements have enabled one to study the QD trionic fine structure[20,21] in high detail. Further investigations have also demonstrated coherently manipulated charged QD states.[18] In this paper we provide direct evidence for the cascaded emission from a charged biexciton 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 QD's 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 three-dimensional (3D) confined individual In(Ga)As quantum dots (height 1–2 nm, width 15–20 nm) within a thin ternary wetting layer [1 monolayer (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 of individual QD's, 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-diam mesa.

Our experiments have been performed on a combined low-temperature (4 K) microphotoluminescence (μ-PL) system and a Hanbury-Brown-Twiss- (HBT-) type photon correlation setup for investigations of 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 τ were measured by a multichannel analyzer. For quasiresonant (*p*-shell) or off-resonant (above GaAs barrier) continuous-wave (cw) excitation a Ti:sapphire laser was used. For pulsed autocorrelation measurements, the laser system was used in its picosecond mode ($\Delta t_{pulse} \approx 2$ 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.

Figure 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 and 1.3372 eV which exhibit nearly resolution-limited linewidths of 55 ± 10 and 75 ± 10 μeV, re-

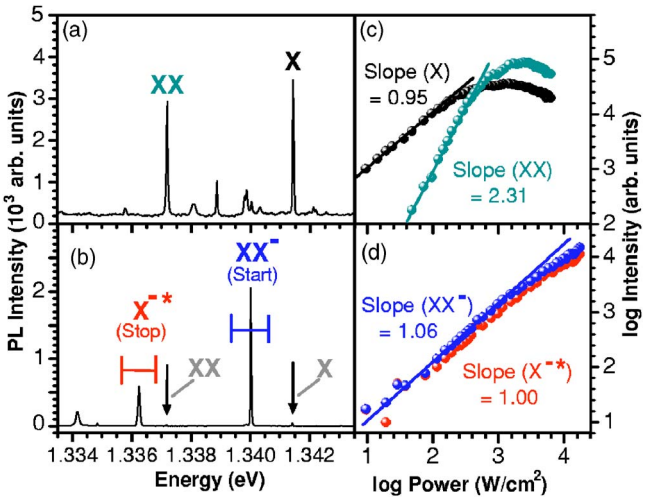

FIG. 1. (Color online) (a) μ-PL spectra from a single (In,Ga)As QD as observed for off-resonant (i.e., above-barrier) cw laser excitation. (b) Spectra taken on the same QD under quasiresonant (p-shell) excitation revealing PL lines being addressed to charged complexes. Horizontal markers: filter bandpass for photon correlation experiments. (c),(d) cw power dependence of neutral and charged QD states under off-resonant (c) and quasiresonant (d) excitation.

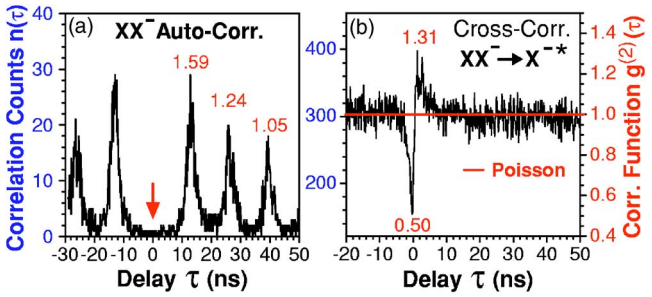

FIG. 2. (Color online) (a) Photon autocorrelation measurement under ps pulsed quasiresonant excitation (E_{exc}=1.407 eV) as obtained from the 1.340-eV PL line [see Fig. 1(b)]. Numbers: Poisson-normalized peak areas. (b) Cross-correlation trace obtained from the 1.3400-eV (XX^-: "start") and 1.3362-eV (X^{-*}: "stop") PL lines in Fig. 1(b).

spectively. As is depicted in Fig. 1(c), the PL of the two lines reveals a linear (slope=0.95±0.05) and superlinear (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 also supported by the observed line spacing of $\Delta E_{X\text{-}XX}$ =4.2±0.1 meV which is consistent with the biexciton-exciton Coulomb exchange energy range for those dots.

As is shown in Fig. 1(b), the emission spectrum completely changes when the exciting laser is tuned energetically into the QD p-shell (E_{exc}^{res}=1.4068±0.0004 eV, P_{exc} =1.9±0.1 kW/cm^2). This quasiresonant excitation reveals the onset of a new set of almost background-free narrow intense lines at 1.3400 eV (50±5 μeV) and 1.3362 eV (80±5 μeV) and a weak 1.3342 eV (110±10 μeV) third signature, all of which appear shifted to lower energies with respect to the strongly suppressed X and XX PL (marked by arrows).

Under p-shell pumping, power-dependent μ-PL series on the two dominant lines [see Fig. 1(d)] revealed an almost linear response for low and moderate densities together with the onset of saturation in the high power limit. It is worth noting that under these quasiresonant conditions a power dependence different from the off-resonant case [see Fig. 1(a)] should indeed be expected.

The assumption of a linear and quadratic power dependence for the X and XX lines, respectively, is based on a simple rate-equation model. Among other assumptions this requires an incoherent regime for the excitation. While this might be approximately the case for above-barrier excitation due to the continuum of states, this cannot be expected for resonant (p-shell) pumping. On the contrary, even Rabi flopping has been reported by Santori *et al.*[22] in this case. Although Rabi flopping is not observed explicitly in our experiment, we cannot expect that rate equations would be applicable for resonant p-shell excitation. Under the conditions of direct pumping into the discrete excited QD states followed by fast relaxation into lower available s states an increase of their occupation proportional to the generation rate of carriers with the laser power seems more intuitive due to the lack of a continuum.

In our experiments the weak 1.3342-eV third PL signature's tendency remained unresolved due to insufficient signal strength. Therefore, the following analysis will focus on the two major lines which we assign to the radiative decay of the *charged biexciton* complex ($1e^2 2e^1 1h^2 \mapsto 1e^1 2e^1 1h^1$) and the consecutive *trion* recombination from its *excited* state ($1e^1 2e^1 1h^1 \mapsto 2e^1$), respectively, as will be demonstrated below.

In order to investigate the photon emission statistics of the observed PL lines we have performed photon autocorrelation measurements using the HBT setup. As an example, the result obtained from the intense XX^- peak (1.3400 eV) under quasiresonant pulsed excitation (3.82 kW/cm^2, averaged) is shown in Fig. 2(a). The given numbers correspond to the normalized correlation peak areas with respect to the Poisson level $N_1 N_2 \Delta t_{laser} \Delta t_{int}$ under consideration of the detector count rates, the laser pulse period, and the total integration time. Multiple-photon ($\geqslant$2) emission is found to be fully suppressed on this PL line which manifests itself in the absence of the zero delay (τ=0 ns) coincidence peak. This behavior demonstrates almost perfect (background-free) triggered single-photon generation from an individual QD.

Furthermore, in Fig. 2(a) correlation peaks at delay times $\tau = nT_{laser}$ ($n = \pm 1, \pm 2, \ldots$) corresponding to correlation events of photons following subsequent laser excitation cycles T_{laser}=13.12 ns reveal a clear *bunchinglike* behavior. In other words, we observe a higher probability to detect another XX^- photon in the next (previous) excitation cycle than in later (earlier) cycles. Such an effect has been reported recently on similar InAs QD's only under the condition of *pure quasiresonant* pumping into an excited state[5,22] and is

interpreted as a fast "blinking" process between the neutral- and charged-state configurations 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. It is important to note that this strongly suggests a residual charging of the QD—e.g., by ionization of adjacent impurities[23]—which supports our preliminary assignment of the PL signatures given in Fig. 1(b). Applying a fit function[22] of the type $g^2(\tau)=1+g_1\exp(-\tau/\tau_{blink})$ for discrete values $\tau=nT_{laser}$ to the Poisson-normalized correlation peak areas of the data in Fig. 2(a), we have extracted the parameters $g_1=1.67\pm0.14$ for the bunching amplitude and $\tau_{blink}=12.8\pm2.0$ ns for the blinking time constant. The short time scale of this effect is consistent with the generally expected behavior for a regime of excitation approaching the saturation level.[22]

The assignment of the observed PL under quasiresonant excitation to charged QD carrier complexes is supported by a simple model scheme: In the absence of laser excitation a doping-related or 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. Under *p*-shell excitation, electrons and holes are injected pairwise into the QD which can consecutively relax into available lower *s* states. Therefore, the creation of odd-number charge states [electron-hole (e-h) pairs plus an extra electron] should be favored. In contrast to this, for above-barrier pumping the effect of *photodepletion*[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 electrons are also partly trapped by the impurity centers. Under these conditions, the radiative decay of neutral X and XX complexes is predicted.

In addition, we have performed photon *cross*-correlation measurements under quasiresonant cw excitation in order to identify the temporal order of the emission between the observed XX^- and X^{-*} lines. Figure 2(b) shows the corresponding correlation histogram as measured $n(\tau)$ (left axis) together with its Poisson-normalized $g^{(2)}(\tau)$ representation (right axis). 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 setup, a pronounced signal asymmetry was observed in the vicinity of zero delay. The antibunching $g^{(2)}(\tau)\to0.5$ for $\tau<0$ together with the observed bunching behavior $g^{(2)}(\tau)\to1.3$ at positive delays τ 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 emission spectra of those QD's have been computed using a full configuration interaction (FCI) procedure.[26,27] For the material parameters given in Ref. 27, which are typical for InGaAs quantum dots, the X^{-*} 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 is shifted below the trion ground-state emission line due to the (charged) biexciton binding energy. In the case of the *excited* trionic line the exchange interaction between the *s*-shell and *p*-shell carriers overcomes the biexcitonic binding energy and reverses the order of the lines. This can be understood by considering the diagrams in Fig. 3(a). One *s* level and two *p* levels are present in each band. All levels are spin degenerate and, assuming cylindrical symmetry for the QD in the wetting-layer plane, the two *p* levels are degenerate, too.

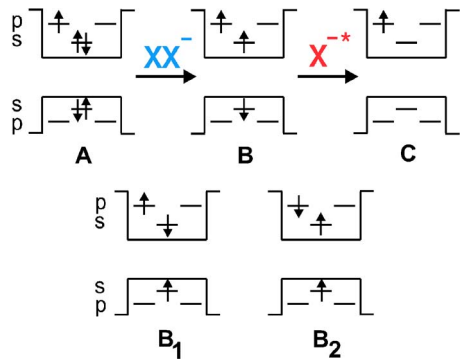

FIG. 3. (a) Scheme of the $XX^-\to X^{-*}\to e$ cascade: Starting from the charged biexciton ground state *A*, an excited charged exciton state *B* is reached by a e-h recombination process. The subsequent recombination step leads to an excited dot state *C* with only one electron. (b) Trion configurations with opposite spins for the *s* and *p* electrons.

The eigenstates provided by the FCI calculations are combinations of many configurations. In Fig. 3(a) only the dominant configurations of the states taking part in the cascade are shown. Their high participation ratio (>96%) allows us to estimate the corresponding energies according to the approximation scheme discussed in Ref. 27. For the energy of the initial configuration *A* this leads to $E_A=2E_s^X+\varepsilon_p^e-X_{sp}$—i.e., the sum of two *s*-exciton energies E_s^X, the one-particle *p*-shell electronic energy ε_p^e, and the (attractive) exchange energy X_{sp} between *s* and *p* electrons with parallel spin. Similarly, $E_B=E_s^X+\varepsilon_p^e-X_{sp}$ and $E_C=\varepsilon_p^e$. For the recombination of the first electron-hole pair, in fact two possible decay paths exist which lead to a final charged exciton state including either two electrons with identical spin orientations [configuration *B* in Fig. 3(a) which will be discussed first] or two electrons with opposite spins [configuration B_1 in Fig. 3(b) which is discussed below].

Strictly speaking, the additivity of different energy contributions leaves out some configuration-interaction effects. One of them is the biexcitonic binding energy, which can be accounted for only by the interaction of the configurations of Fig. 3(a) with the other possible arrangements of the carriers on the available states. Considering only the *s* and *p* shells and based on the material parameters of Ref. 27 one obtains from FCI calculations a biexciton binding energy of ≈2 meV for the neutral biexciton and ≈1.2 meV for the charged one.

Including this configuration-interaction effect, the energy of configuration *A* reads $E_A=2E_s^X-\Delta+\varepsilon_p^e-X_{sp}$ with the charged biexciton binding energy Δ. As a result one finds the following emission energies:

$$E_{XX^-} = E_A - E_B = E_s^X - \Delta,$$

$$E_{X^{-*}} = E_B - E_C = E_s^X - X_{sp}. \quad (1)$$

The difference in the positions of the two lines is given by $X_{sp} - \Delta$. The exchange integral X_{sp} (in our case ≈ 4.9 meV) is significantly larger than the biexcitonic binding energy Δ, which explains the energetic positions $E_{X^{-*}} < E_{XX^-}$.

We would like to point out that the biexciton binding energy being smaller than the *s*-*p* exchange energy is not a matter of special parameters. While our example, based on the material parameters of Ref. 27, uses interaction matrix elements typical for the InGaAs quantum dots, there are more general arguments showing that the discussed situation is expected rather than coincidental. It is illuminating to compare the QD biexciton problem with the hydrogen molecule (Heitler-London) theory. In the latter, the positive charges keep an optimal distance from one another and allow the negative ones to take the space between. In the quantum dots, due to the discrete one-particle spectrum, no such geometrical flexibility is allowed. The carriers take preferentially the lowest available states, in which all the charges are superimposed. In the limiting case of identical envelopes (accurate within a few percent as far as Coulomb matrix elements are concerned; see Refs. 28 and 29) the exciton is not only globally but also locally neutral, so that the two excitons do not interact at all, which would lead to a vanishing biexciton binding energy. The corrections to this picture, contributions of higher-state configurations, allow for a rearrangement of the charges. But the energetical price to be paid keeps these admixtures small, and therefore the biexciton binding energy is small. This is a common result of different calculations of QD spectra, based on various parameters or models for the confinement potential.[30–32] The picture applies even more to the case of the charged biexciton, because the presence of the additional carrier limits the available higher states. Therefore it is not a numerical accident but rather a physical fact that one obtains, for a whole range of reasonable parameters, the same conclusion: the *s*-*p* exchange energy overtakes the (charged) biexciton binding energy and therefore the X^{-*} line appears at an appreciably lower energy.

To complete the discussion, we point out that the initial charged biexciton state is actually fourfold degenerate, since four states are available to the *p* electron. Moreover, the *s* electron left in the excited trion may not necessarily have the same spin as the *p* electron. Such a situation is shown in configuration B_1 of Fig. 3(b). In this case the above argument seems to fail, since the exchange integral apparently plays no role anymore, though the FCI calculation shows that this is *not* the case. Actually, configuration B_1 alone is not an eigenstate of the problem. It gets mixed with configuration B_2 (which cannot be directly obtained from the radiative decay of *A*), thus resulting in a state with exactly the same energy as state *B*. This is by no means a coincidence: The electrons in configuration *B* have the spin state $S^e=1$, $S_z^e=1$ and the sum of B_1 and B_2 corresponds to $S^e=1$, $S_z^e=0$. These states are necessarily degenerate in an eigenvalue problem which conserves both the electron and hole spin.[33]

By reducing the symmetry of the problem some of the degeneracies are removed and additional lines may appear in PL. For example, if it is the rotation symmetry that is broken, the two *p* states are not equivalent anymore. One then expects different one-particle energies for the "left" and "right" *p* states and also the exchange integral would take two different values. By examining Eq. (1) one sees that both emission energies are insensitive to the position of the *p* level while only the X^{-*} line is sensitive to the change in the X_{sp} integral. In this case it is only the lower line which becomes split by the symmetry lowering. This might be a possible explanation for the appearance of a third line below X^{-*}. However, since it was currently not possible to use the weak signal of this line for correlation measurements, its identification has to be left for further investigations.

In addition to the experiments discussed before, we have also tested the possibility to generate polarization-correlated photon pairs by the $XX^- \mapsto X^{-*}$ cascade under investigation. This effect is connected with the splitting of the involved excited trion ($S=1$) triplet state due to electron-hole-exchange interaction and would lead to a splitting of both PL lines of the cascade (since the initial XX^- and the final state are not split). Surprisingly, this effect was not seen even within our high limits of spectral resolution so that in this case the exchange splitting is apparently not strong enough.

As a consequence, polarization correlations between consecutively two emitted photons could not be resolved. Together with the effect of a blocked *p*-shell electron relaxation in the excited trion configuration (see preceding discussion), these data might allow us to gain insight into the spin dynamics of these systems. For higher-dimensional systems (see, e.g., Ref. 34) the fastest relaxation is known to occur for the *hole* spin, followed by that of the *exciton*, whereas the *electron* spin takes longest to flip. This hierarchy seems to be maintained also for the QD structures under study: The fact that emission from the excited trion complex is observed indicates a considerably longer spin-flip time for the *p*-shell electron than for the electron-hole recombination. Since time-resolved PL on single QD's has revealed a decay time of ≈ 1 ns, this gives a lower limit for the electron spin-flip time in the excited state. On the other hand, the absence of a photon polarization-correlation[35] in the cascade might be explained only by an *s*-shell exciton spin-flip process being significantly faster than the radiative lifetime ($\tau_{flip} \ll 1$ ns).

In conclusion, the emission from charged exciton and biexciton carrier complexes has been observed under the condition of pure quasiresonant excitation. Besides the capability of background-free triggered single-photon generation, the cascaded nature of these decay channels was directly demonstrated in 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 have been found, thus reflecting a long-lived excess electron spin configuration.

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