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Photon correlation spectroscopy of a single quantum dot

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We report photon correlation measurements that reveal unique signatures of biexciton and charged exciton emissions in a single self-assembled InAs quantum dot. Biexciton emission exhibits both bunching and antibunching under continuous-wave excitation and only antibunching under pulsed excitation. Cross correlation between biexciton and single-exciton emissions reveals highly asymmetric features due to cascaded emission. Under our excitation conditions, no polarization correlations between the exciton and biexciton emissions were observed.

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It is by now widely accepted that various quantum dot (QD) structures exhibit features in transport[1,2] or optical spectroscopy[3–5] that indicate full three-dimensional confinement of carriers. Identification of QD's as artificial atoms has been strengthened by the recent observation of strong photon antibunching in single-exciton emission,[6,7] which is the typical signature of an anharmonic quantum system: after a photon is emitted from a single two-level (anharmonic) emitter, the system is necessarily in the radiatively inactive ground state and a second photon cannot be emitted immediately after the first one. Even though the coherence properties of QD single-exciton emission closely follow those of atoms, the overall spectral features of single QD's are significantly more complicated. Since the size of QD's is roughly two orders of magnitude larger than those of atoms, multiparticle excitations give rise to emission peaks with energies comparable to that of a single exciton. Of primary importance in QD spectroscopy is the biexciton state, which corresponds to a doubly-excited QD with completely filled lowest electron- and hole-energy levels. When the biexciton state decays by radiative recombination, the final state is a single-exciton state and the generated photon is shifted as compared to the single exciton emission due to Coulomb interaction between the carriers. Biexciton emission in QD spectroscopy has been traditionally identified using the (quadratic) pump-power dependence of the corresponding peak.

In this paper, we demonstrate that photon-correlation measurements provide a powerful tool for characterizing the multiexciton spectral features of QD's. Our measurements provide a strong support for the identification of a biexciton emission peak, by demonstrating its strong correlations with the subsequent single-exciton emission. We observe that biexciton intensity autocorrelation exhibits bunching together with antibunching or only antibunching under continuous-wave (cw) excitation depending on the excitation level. In contrast, we find strong antibunching under pulsed excitation. The large difference between the levels of antibunching under continuous wave and pulsed excitations points out to the importance of excitation mechanism and the role of free carriers in QD physics. The lack of polarization correlation between biexciton and single-exciton emissions indicates that spin dephasing is likely to play a key role under nonresonant excitation. We also observe that a third emission peak in QD spectra exhibits strong correlations with both exciton and biexciton fluorescence: we argue that these correlation signatures suggest the identification of this additional line as a charged-exciton emission.

Our self-assembled InAs QD's were grown by molecular-beam epitaxy using the partially covered island technique.[8] Growth resulted in typically lens-shaped QD's with a base diameter of 40–50 nm and a height of 3 nm, having their single-excitonic emissions between 925 nm and 975 nm in the spectrum. In our sample, the QD's were embedded in the center of a 200-nm-thick GaAs microdisk structure located above a 0.5-μm-thick $Al_{0.65}Ga_{0.35}As$ post. The diameter of the disks was 5-μm and the average number of QD's within the disks was less than one. Details of the microdisk processing can be found elsewhere.[9] Our experimental setup consisted of a combination of a low-temperature diffraction-limited scanning optical microscope and a Hanbury Brown and Twiss (HBT) setup[10] for photon-correlation measurements. The sample was mounted in a He-flow cryostat and cooled to 4–7 K. The QD's were optically excited either with a continuous-wave diode laser (operating at 785 nm), a continuous-wave Ti:Sa laser (operating at 760 nm) or a mode-locked Ti:Sa laser (82 MHz, 250 fs, operating at 790 nm), generating electron-hole pairs in the GaAs barrier layer, which are subsequently captured by the QD's within a short timescale[11] (less than 35 ps). The same microscope objective (NA=0.55) mounted outside the cryostat was used to both excite the QD's (diffraction limit: ~ 1.7 μm) and collect the emitted fluorescence light. The collected light was spectrally filtered either by a 0.5 m monochromator or narrow-band interference filters (full width at half maximum is 0.5 or 1 nm) before being detected in the HBT setup, consisting of a 50/50 beamsplitter and two single-photon-counting avalanche photodiodes (APD's).[7] The time resolution of the HBT setup was 420 ps. Under our experimental conditions with detection efficiency of $\approx 0.1\%$, the measured photon-pair distribution directly yields the normalized (second-order) intensity autocorrelation (coherence) function $g^{(2)}(\tau)$

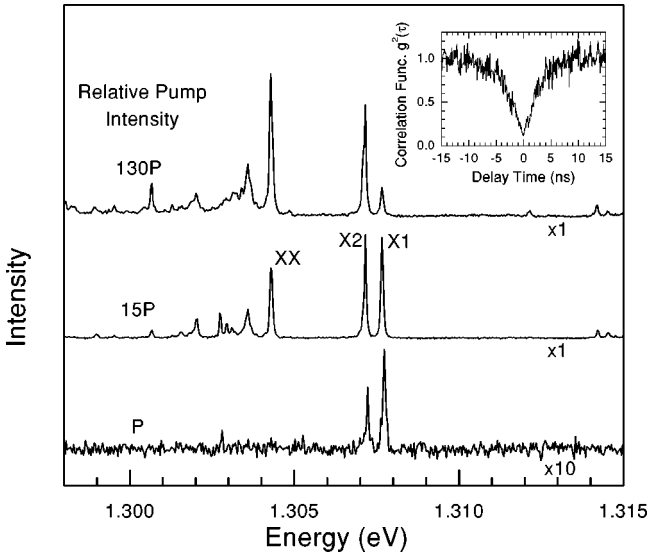

FIG. 1. PL spectra for the quantum dot studied in this paper for various powers of the cw diode laser at 785 nm. Indicated are the three peaks that we focus on. With increasing powers, the dominant line is successively X1, then X2, then XX. All lines saturate and then decrease with increasing power. Inset, photon-correlation measurement carried out using the X1 emission, showing strong antibunching.

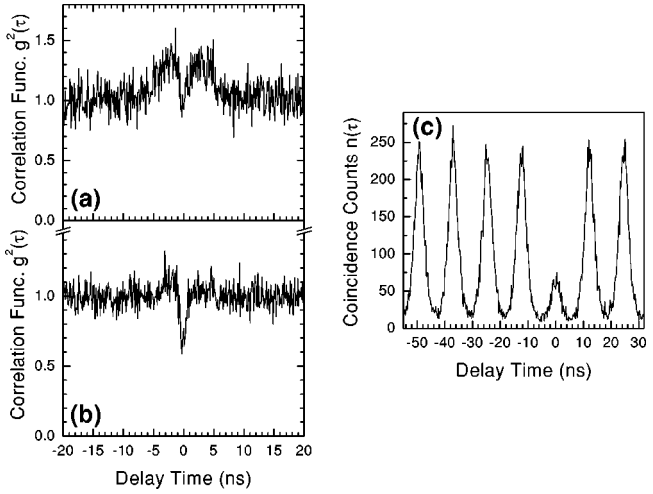

FIG. 2. Measured second-order correlation function of the XX line. Under cw excitation (Ti:Sa laser, 760 nm) (a) an antibunching dip ($g^{(2)}=0.95$) together with a bunching peak ($g^{(2)}=1.4$), and (b) an antibunching dip ($g^{(2)}=0.6$) are visible at X1 emission intensities 0.7 and 1.0 of the (exciton) saturation level, respectively. (c) Under pulsed excitation (82 MHz, 250 fs, at 790 nm), the relative area of the central peak compared to the other peaks is 0.3, indicating strong antibunching in contrast with the cw results.

$=\langle :I(t)I(t+\tau):\rangle/\langle I(t)\rangle^2$. Here :: denotes normal ordering and $I(t)$ is the intensity operator.

Figure 1 shows the power-dependent photoluminescence (PL) spectra of the single QD that we analyze. At low pump powers, the single-exciton emission peak (X1) dominates the spectrum. At higher pump powers, we observe that two other peaks become dominant: among these, the lower energy one (XX) has an energy (red) shift of 3.5 meV from X1 and its intensity has a quadratic dependence on pump power, these are typical signatures for biexciton emission in self-assembled InAs QD's. The third peak (X2) is red shifted from the single-exciton peak by about 500 μeV. All three emission peaks are resolution limited at 70 μeV, and none of them is polarized. To ensure that X1 originates from a single QD exciton emission, we have carried out photon autocorrelation measurements where both APD's were illuminated by X1: under cw and pulsed excitation, X1 emission was found to exhibit perfect antibunching (Fig. 1 inset) and single-photon source operation,[12,13] respectively. We have also performed time-correlated single-photon counting experiments on X1, X2, and XX emissions to measure their lifetimes.[14] Those measurements were performed in the very low excitation regime where the decay times of the resulting spectra were determined by the lifetimes of the corresponding emissions.[15] From the measured spectra we deduced lifetimes of 3.6 ns, 3.7 ns, and 2.6 ns for the X1, X2, and XX emissions, respectively. The resultant ratio of $\tau_{X1}/\tau_{XX}=1.4$ is consistent with exciton and biexciton lifetime measurements performed on CdSe/ZnSe QD's.[15] We also note that, due to the different photonic environment created by the microdisk, the single-exciton lifetime we observe (3.6 ns) is larger than typical lifetimes measured ($\sim$1 ns) in bulk QD samples.[7]

Since the QD spectrum is anharmonic, it could be argued that the measurement of $g^{(2)}(\tau)$ for the XX line will also exhibit simple antibunching, as has been observed for single-exciton emission. Figures 2(a) and 2(b) show $g^{(2)}(\tau)$ for the XX emission under cw excitation, at pump powers corresponding to X1 emission intensities that are 0.7 and 1.0 of the (exciton) saturation level, respectively. Both curves, obtained using a 0.5-nm interference filter, exhibit antibunching [$g^{(2)}(0)=0.95$ in Fig. 2(a), $g^{(2)}(0)=0.6$ in Fig. 2(b)] with similar decay times of 1 ns. The curve in Fig. 2(a) also exhibits bunching ($g^{(2)}=1.4$) that decays with a decay time of 3.5 ns. Bunching here originates from the fact that the detection of a photon at the biexciton transition results in the projection of the QD wave function onto the single-exciton state X1. When the average occupancy of X1 in steady state is lower than unity, postmeasurement state has higher occupancy in the single-exciton state than premeasurement state, and is more likely to result in reexcitation of the biexciton state. An analysis of the QD dynamics using three-level rate equations indicates that $g^{(2)}(\tau)$ should indeed exhibit bunching that decays in a timescale determined by the single-exciton lifetime of 3.6 ns, which is in agreement with the experimental result [Fig. 2(a)]. This analysis also predicts that antibunching at $\tau=0$ should turn into bunching in a timescale determined by the biexciton lifetime in the low excitation regime.

When we replace the 0.5-nm interference filter by a 1-nm filter, we cannot observe antibunching in biexciton autocorrelation measurements, even though strong bunching feature around $\tau=0$ persists. This finding highlights the importance of broadband background radiation at biexciton energy. The fact that we continue to observe strong bunching indicates that this background is correlated with exciton emission, much similar to the biexciton emission itself. The strength of

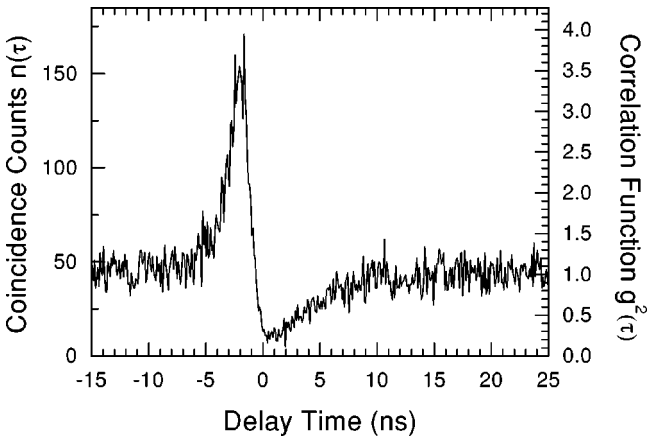

FIG. 3. Cross-correlation function between X1 and XX emissions under cw diode laser excitation at 785 nm. For the chosen pump intensity, X1 line is well below saturation. The signal from X1, filtered by a 0.5-nm interference filter, is sent to the start APD, while the XX line, filtered by a 1-nm filter, is sent to the stop APD. The positive correlation ($g^{(2)}=3.4$) for $\tau<0$ followed by the negative correlation ($g^{(2)}=0.2$) for $\tau>0$ is an evidence for the cascaded emission of photons.

biexciton antibunching depends on the pump-laser wavelength but not on its intensity, provided that the quantum dot is not well above saturation. This result is inconsistent with our expectation that biexciton antibunching will be strongly influenced by free carrier density. On the other hand, the significantly stronger biexciton antibunching that we observe under pulsed excitation [Fig. 2(c)] suggests that the free carriers may still be playing a role: we remark that under pulsed excitation, free carriers recombine in a time scale that is much faster than the biexciton radiative recombination time,[16] and, therefore, their influence on biexciton dynamics is expected to be minimal.

Cross-correlation measurements complement the identification of the biexciton emission: since X1 population is enhanced as a result of the detection of an XX photon, strong correlations between the X1 and XX emissions can be expected. Figure 3 shows such a photon cross-correlation measurement, obtained by illuminating the start APD by the X1 emission and stop APD by the XX emission. The depicted quantity here is $\tilde{g}^{(2)}(\tau)=\langle :I_{XX}(t)I_{X1}(t+\tau):\rangle / [\langle I_{X1}(t)\rangle\langle I_{XX}(t)\rangle]$, where $I_{X1}(t)$ and $I_{XX}(t)$ are the intensities of the X1 and XX emissions, respectively. Remarkable features of this cross correlation include strong antibunching for $\tau>0$ and strong bunching for $\tau<0$ with a close to resolution-limited transition between the two regimes. For $\tau>0$, suppression of a joint X1 and XX event arises from the fact that following the detection of an X1 photon, which projects the QD onto its ground state, detection of an XX photon is very unlikely. Strong bunching for $\tau<0$ follows from the fact that detection of an XX photon projects the QD onto the X1 state, as discussed earlier. Asymmetry in X1 and XX cross-correlation measurements have been recently reported.[17,18] The signature depicted in Fig. 3 proves that the XX emission arises from the decay of the biexciton state into the single-exciton state. The strong antibunching in cross correlation is yet another indication that the additional broadband radiation is correlated with the X1 emission.

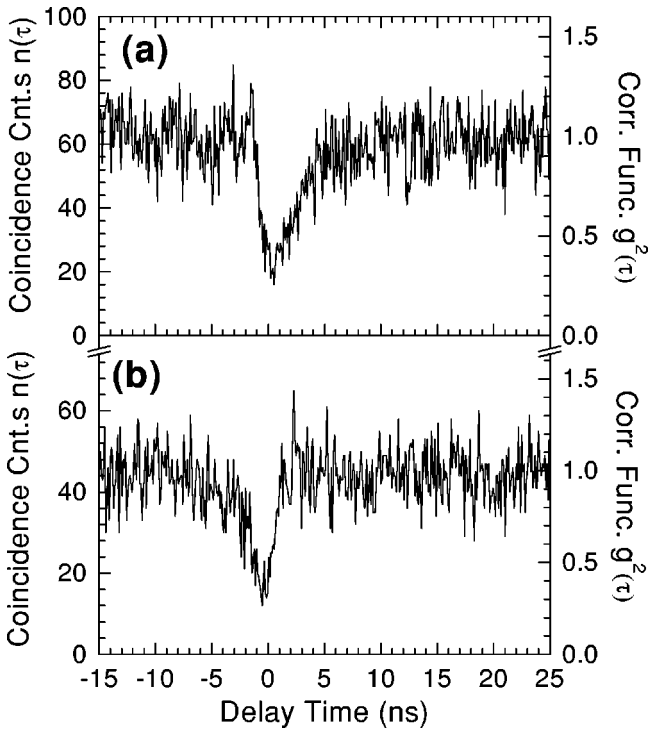

FIG. 4. Cross-correlation measurements under cw diode laser excitation at 785 nm. (a) The X2 fluorescence is sent to the start APD while XX is sent to the stop APD. The absence of bunching demonstrates that XX emission does not populate the X2 state. (b) The X2 emission is sent to the start APD and the X1 emission to the stop APD. The antibunching dip shows that both transitions stem from the same QD. The asymmetry in the dip indicates a faster recovery for the X1 state after a X2 detection.

Having identified the two principal lines in QD spectrum, the next natural question is whether photon-correlation spectroscopy can tell us anything about the origin of the X2 emission. The cross correlation between the X2 and XX emissions only shows antibunching [Fig. 4(a)], indicating that while those emissions arise from the same QD, the radiative decay of the biexciton state does not populate the X2 state. From the pump-power-dependent PL spectra (Fig. 1), it can be seen that the X2 emission has a stronger pump-power dependence than X1 but saturates earlier than the XX line. This may already suggest an identification of X2 as a charged-exciton (trion) line despite the fact that X2-X1 energy difference is smaller than typical reported values. To provide further evidence, we have carried out cross-correlation measurements between the X2 and X1 emissions $[\overline{g^{(2)}}(\tau)]$ where the start and stop APD's were illuminated by the X1 and X2 lines, respectively. The resulting X1-X2 cross-correlation function [Fig. 4(b)] clearly shows asymmetric antibunching with $\overline{g^{(2)}}(0)=0.3$, which proves once again that the two lines originate from the same QD. The asymmetry with fast recovery for $\tau>0$ is expected if X2 arises from a charged exciton: the postmeasurement state of charged-exciton emission is a singly-charged QD. We would expect single-charge injection into the QD to be much faster

than triple-charge injection, which in turn determines the recovery time for $\tau<0$. Given that the X2 emission of this QD is stronger than what we typically see in other QD's, we could envision the presence of an acceptor or donor impurity that increases the relative intensity of charged-exciton emission. Presence of carbon in these samples is well known. Under resonant (p-shell) excitation condition we only observe X2 emission, as would be expected from a doped QD. We have carried out experiments on other QD's and observed identical asymmetric antibunching signatures in cross correlation between the fundamental exciton line and a secondary redshifted line.

Finally, it has been predicted that the radiative decay of a single QD biexciton state will result in polarization-entangled-state generation.[19] To observe such polarization correlations, we have measured the polarization dependence of the X1-XX cross correlation. Under cw excitation, we have seen no evidence for polarization correlations. We believe that spin decoherence that has been observed to occur in nanosecond timescales for these QD's under nonresonant excitation is responsible for the lack of polarization correlation.[20]

In summary, we have used photon auto- and cross-correlation measurements to identify dominant spectral features of a single QD, and characterize the recombination dynamics under various excitation conditions. Given the difficulty of accurate theoretical calculations and the richness of the QD spectra that differs significantly from one QD to another, we believe that the techniques described here will be invaluable in understanding individual QD's. Further experiments under different excitation conditions are needed to understand the polarization correlations and eventually for the generation of entangled-photon states.

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