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Single-photon emission from exciton complexes in individual quantum dots

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Applications in optical quantum information technology require a new type of light source able to emit exactly one photon periodically with minimal timing jitter. We investigate the photon emission statistics of different electron-hole recombination processes in a single photo-excited semiconductor quantum dot. We demonstrate single photon emission from each of these exciton complexes, revealing the biexciton state to produce single photons with significantly less jitter in their emission time than the single exciton state.

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The statistical nature of photon emission from conventional light sources results in random fluctuations in the number of emitted photons in a given time interval. For instance, the distribution in the number of photons in a laser pulse obeys Poissonian statistics. There is currently much interest in developing a new type of light source, which emits exactly one photon at regularly spaced time intervals. This could be useful in quantum cryptography, for instance, which would allow a cryptographic key to be formed from bits encoded upon single photons transmitted along an optical fiber.[1] By using single photons, the sender and intended recipient are able to guarantee the security of their key, since quantum mechanics dictates that measurement by a third party will inevitably produce a detectable change to the encoded single photons. In the absence of single photon source, practical demonstrations of quantum key distribution have used a highly attenuated pulsed laser diode. However, it has been shown that the multiphoton pulses, which are inevitable when using laser light, render the technique insecure over fiber lengths of more than a few tens of kilometers.[2] Secure quantum key distribution over longer distances therefore requires the development of a true single-photon source. Indeed such a source will be an essential building block for a great many applications in photonic quantum communications[1] and computing.[3]

It has been long realized that the resonance fluorescence of a single two-level atom should display photon antibunching, since the emission of a photon returns the atom to its ground state.[4] This was first demonstrated experimentally in the resonance fluorescence of a low density vapor of Na atoms[5] and later for a single trapped Mg^+ ion.[6] Since then the photon emission statistics of a range of other quantized, two-level systems have been studied, such as single molecules,[7–11] CdSe/ZnS nanocrystals,[12] and nitrogen vacancy centers in diamond.[13,14]

A quantum dot is often described as the semiconductor analogue of an atom, since the three-dimensional confinement of the electrons results in their energy spectrum consisting of a series of discrete lines. Each of these levels can accommodate just two electrons of different spin, due to the Pauli exclusion principle. Recently, this property of quantum dots has been utilized to demonstrate single photon emission due to single electron-hole pairs photo-excited in a quantum dot.[15,16] Compared to atoms, quantum dots have the advantage of mechanical stability and long lifetime, while also allowing nonresonant laser excitation and possible integration with conventional semiconductor light-emitting technology.

Another possible advantage of quantum dots is that they allow control over the atomiclike emitting species, through confinement of different numbers of electrons and holes within the dot.[17–20] In this paper we study the photon statistics of the emission from the different electron/hole combinations that can be confined in the dot. Through a study of the spectral, temporal, and intensity dependence of the dot emission, we assign transitions due to the biexciton, charged exciton, and charged biexciton, as well as the simple exciton of one electron and one hole. Using a Hanbury–Brown and Twiss correlation experiment we demonstrate the suppression of multiphoton emission from each of these exciton complexes. We find the biexciton transition is actually more favorable for single photon emission than the single exciton due to the much lower jitter in its emission time.

The sample studied was grown by molecular beam epitaxy on a GaAs substrate, and consists of a self-assembled quantum dot layer formed by depositing 1.7 monolayers of InAs onto a 500 nm GaAs buffer, followed by a 300 nm GaAs cap. Single quantum dots were isolated by wet etching 0.8 μm mesas on the wafer surface. The sample was placed in a cold finger liquid helium cryostat at 5 K, and was excited nonresonantly with picosecond pulses from a modelocked titanium-sapphire laser at an energy of 1.55 eV, above the band gap of the GaAs barrier layers. The laser was focused to a $\sim$1 μm spot on the surface of the sample by an infinity-corrected microscope objective lens, which also collimated photoluminescence from the sample. This was dispersed by a grating spectrometer and detected using either a charge-coupled device (CCD) or an avalanche photo diode (APD). The magnification was such that the image of the PL from a mesa was $\sim$100 μm, which allowed spatial filtering of the photoluminescence by adjusting the size of the spectrometer entrance slit. The spectral resolution of our system was $\sim$50 μeV, and the spatial resolution was $\sim$1 μm.

Time-integrated PL spectra for selected excitation powers are presented in Fig. 1. At the lowest laser power of 0.13 nW, two lines are seen: The stronger marked *X* at 1.3748 eV and

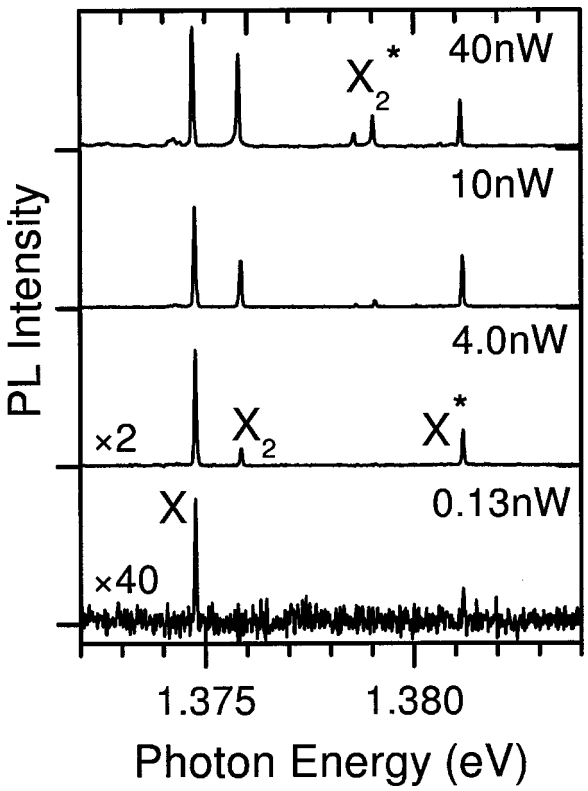

FIG. 1. Photoluminescence spectra of a single quantum dot recorded for different laser excitation powers. The appearance of more lines can be seen as the power is increased.

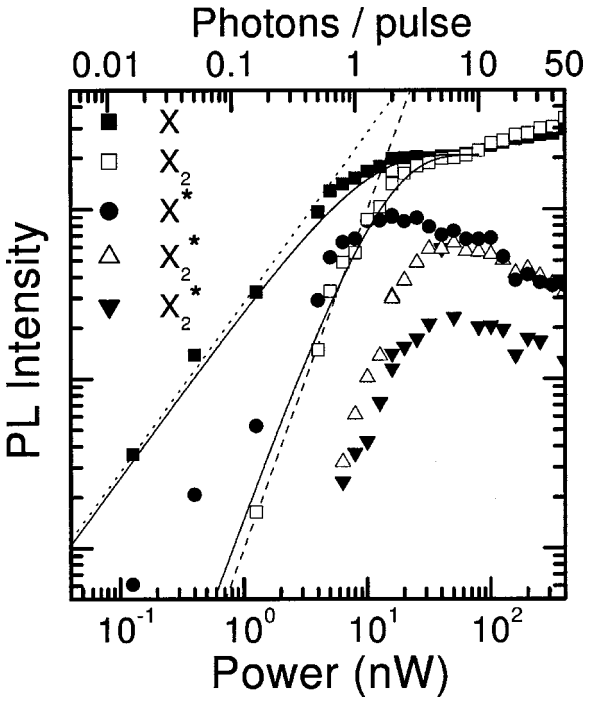

FIG. 2. Integrated intensities of PL lines as a function of laser excitation power. Open and solid triangles represent the higher and lower energy components of the X_2^* doublet. Dotted (dashed) line shows the gradient associated with linear (quadratic) power dependence. The solid lines show calculated power dependence of X and X_2 as a function of photons per pulse absorbed close to the dot.

the weaker marked X^* at 1.3812 eV. As the power is increased to 4.0 nW, a third line (X_2) appears at an energy 1.1 meV higher than that of X. At 10 nW and a pair of lines X_2^* appear at 1.3786 eV and 1.3790 eV. All lines are saturated in intensity at the highest laser power shown of 40 nW, and no other lines are observed at any photon energy up to that of the wetting layer emission. The fact that no groups of lines appear to higher energy is strong evidence for the existence of only one pair of electron and hole levels in the quantum dot. Other PL studies on single quantum dots[18,20,21] have shown the appearance of excited state emission around 30–60 meV to higher energy of the lines seen at lowest powers. However, the dots studied here have a smaller size, as demonstrated by their higher emission energy. It is therefore reasonable to conclude that the energy-level spacing in our dots is much larger than found in other work, resulting in only a single confined electron or hole level. We observed a qualitatively similar PL line structure to Fig. 1 from other mesas containing a single dot.

To identify the exciton complexes responsible for the observed emission lines, we studied their integrated intensities as a function of laser power, as plotted in Fig. 2. The intensity of lines X and X^* show an approximately linear power dependence, with exponents of 0.94 ± 0.04 and 1.09 ± 0.09, and saturate at approximately the same power of 8 nW. Therefore X and X^* are attributed to quantum dot configurations containing only a single electron-hole pair. The onset of emission from X_2 and X_2^* occurs at higher laser powers than for X and X^*, and the intensity of the emission increases with approximately quadratic power dependence, with fitted exponents of 1.95 ± 0.12 and 2.02 ± 0.15 respectively. This strongly suggests that X_2 and X_2^* are due to emission from the quantum dot containing two electron-hole pairs. The fact that excited states are not observed rules out the possibility of attributing the emission to dots containing three or more excitons. In addition, the higher and lower energy components of the X_2^* pair have an almost constant intensity ratio. They are therefore regarded as a doublet from different configurations of the same multicarrier complex.

At high excitation powers >80 nW, there is a redistribution of the emission intensity from X^* and X_2^* towards X and X_2, demonstrating that emission from X and X_2 is a complementary process to emission from X^* and X_2^*. This is supported by the similar intensities at high laser power of X with X_2, and X^* with X_2^*. The line structure for X and X_2 is very different to that for X^* and X_2^*, which makes it difficult to attribute the emission to two different dots within the same pillar. This conclusion is also supported by emission from other pillars that seem to contain only a single quantum dot, which show qualitatively similar PL spectra and dependence on laser power, as that shown in Fig. 1. A more likely possibility is that the quantum dot may intermittently capture an excess carrier to allow formation of charged as well as neutral excitons, as reported previously.[17,19,20] The presence of charged biexciton emission (X_2^*) suggests that the quantum dot has either a second confined electron or hole level, although the absence of a triexciton transition rules out the

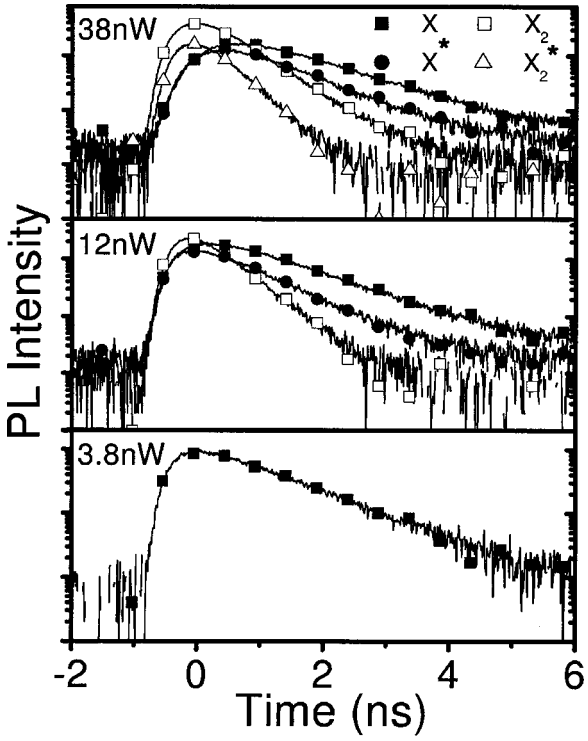

FIG. 3. Time resolved PL measured for each exciton complex at different laser excitation powers. Notice that each complex shows a distinct lifetime which is independent of the laser power. At the higher powers the emission of the single exciton (X) is delayed until after that of the biexciton (X_2). Similarly the charged exciton (X^*) is emitted after the charged biexciton (X_2^*).

possibility that both are bound. Transitions between ground and excited levels are parity forbidden. At high laser powers, it is likely that photo-excited carriers will tend to neutralize the charge trapped in the dots. The four features are thus attributed to the neutral (X) and charged exciton (X^*), and the neutral (X_2) and charged biexciton (X_2^*).

Figure 3 plots the temporal dependence of the emission from the different exciton complexes at different laser powers. At the lowest power of 3.8 nW, only emission from X is measured, displaying a single exponential decay with a lifetime of 1.36 ns similar to that reported previously.[21] The measured rise of the PL is limited by the response of the APD. As the power is increased to 12 nW, the two additional lines X^* and X_2 can be measured, with decay times of 1.07 ± 0.02 ns and 0.59 ± 0.02 ns respectively. The rise of these curves is again limited by the temporal resolution of the system. The PL due to X shows a similar decay time to that at lower power, but the peak intensity of the X PL is delayed by $\sim$0.23 ns relative to its position for the lowest laser power. This is attributed to the time delay associated with the radiative decay of X_2 into X for some of the pulses. At the highest power shown of 38 nW, all lines have reached their maximum intensity, and their temporal characteristics are found to be independent of laser power. In this case the maximum intensity of the X PL is shifted by 0.61 ns relative to that of X_2, as well as that of X for lower laser power. This is in excellent agreement with the measured radiative lifetime of X_2, of 0.59 ± 0.02 ns, which is common with the lifetimes of all exciton complexes studied, is found to be constant as function of power. Similarly, the peak of X^* is shifted by 0.44 ns, and is in agreement with the radiative lifetime of X_2^*, measured to be 0.52 ± 0.05 ns.

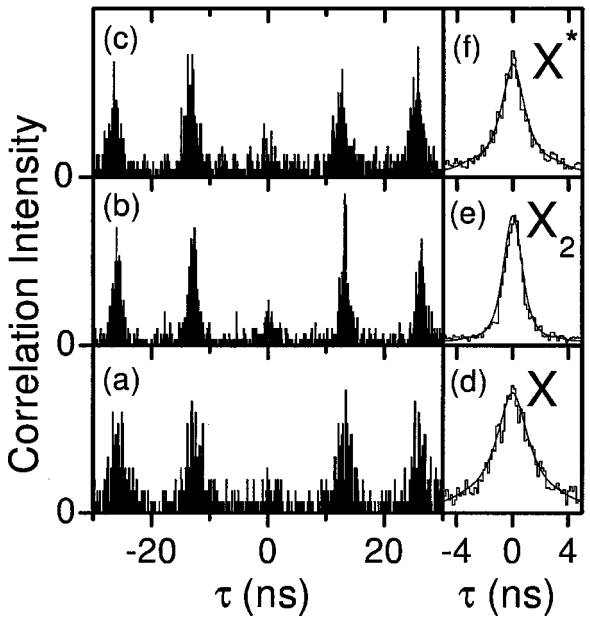

FIG. 4. Second order correlation function measured for emission from the exciton (X), biexciton (X_2), and charged exciton (X^*) are shown in panels (a) to (c). The average peak shape associated with each type of exciton is shown in panels (d) to (f). Smooth lines are calculated by a convolution of time resolved PL decay curves.

The time resolved PL demonstrates that the exciton emission follows that of biexciton, which is to be expected since the biexciton state decays radiatively into the single exciton ($X_2 \rightarrow X + \text{photon}$). Thus the temporal dependence confirms the assignment of these lines from the power dependence of their integrated intensities discussed above. Similarly, emission due to X^* follows that of X_2^*, as expected from the radiative decay of a charged biexciton state into a charged single exciton ($X_2^* \rightarrow X^* + \text{photon}$).

The radiative lifetime of X and X^* are determined to be 1.36 ± 0.06 ns and 1.07 ± 0.02 ns respectively, more than a factor of two longer than the corresponding biexciton. This is attributed to the two possible recombination paths for the two electron-hole pairs in the biexciton. In addition, X^* has a slightly shorter radiative lifetime than X. This may be due to the lack of a dark state for the charged exciton, which allows X^* to be tentatively termed the charged exciton, and X the neutral exciton.

The system was then arranged to measure the second order correlation function, $g^{(2)}(\tau)$, between photons emitted from X, X^*, and X_2 states, using the Hanbury–Brown and Twiss set up. PL excited with a relatively high laser power, so as to saturate the emission intensities, was spectrally filtered by the spectrometer so as to contain just one emission line. This was directed by a 50/50 beamsplitter to two photon counting avalanche photodiodes, and the time delay (τ)

between a count in the two detectors recorded by a time interval analyzer. The second order correlation function is given by the distribution in the measured time delays.

Figure 4 plots the second order correlation function recorded for the X, X^*, and X_2 lines. Each consists of a series of peaks separated by the laser period of 13.0 ns. Notice the strong suppression of the peak around zero time delay for each of the complexes. This is clear evidence for single photon emission from the biexciton and charged exciton complexes in addition to the simple exciton reported previously. The area of the zero time delay peak observed for the X transition suggests a strong suppression of multiphoton pulses by a factor of 15 relative to a Poissonian source of the same efficiency.. We believe that the residual multiphoton pulses derive from stray emission from the buffer and substrate layers of the device and could be reduced by redesign of the sample structure or better spectral rejection.

There are potential advantages in designing single-photon emitting devices around biexcitonic emission from quantum dots rather than the single exciton transition. Since the radiative lifetime of the biexciton is shorter than that of the exciton, by at least a factor of two in these measurements, the maximum possible emission rate from the biexciton state can be higher. Another advantage is a reduction in the timing jitter associated with the uncertainty in the time between photons. This would allow the photon detector used in the application to be gated ''on'' for a shorter time, thus reducing its dark count probability. Figures 4(d)–4(f) show the average peak shape for exciton, biexciton, and charged exciton emission. The full width at half maximum (FWHM) of these measurements directly demonstrates the reduction in jitter of the biexciton emission relative to the simple exciton from 2.79 ns to 1.50 ns. The measured peak widths are in excellent agreement with calculations based on the convolution of time-resolved PL of the corresponding exciton complex, as can be seen in Figs. 4(d)–4(f).

Since triexcitons cannot be confined in these small dots, the temporal evolution of the biexciton PL remains unchanged at higher excitation powers, and the jitter and maximum bit rate are independent of power, unlike that for the single exciton for which we observe a delay at high laser power, or even for a biexciton in a dot with more than one pair of electron and hole levels. This means that the average number of photons emitted by the device per pulse can be much closer to unity, as lower powers are not necessary to reduce the jitter. This has the effect of reducing the number of pulses that contain no photons, and thus increases the emission efficiency.

In summary, we have identified emission lines from an individual quantum dot from laser power dependent and time resolved PL measurements. Time resolved PL measurements show that the radiative lifetime for the exciton complexes studied is constant for all laser powers. The lifetime of the biexciton is measured to be 0.59 ns, a factor of 2.3 times shorter than the lifetime of the exciton. Correlation measurements have shown single photon emission from charged excitons and biexcitons in addition to the simple exciton. The FWHM of the X_2 peaks in the correlation measurement is found to be a factor of 1.9 times smaller than the FWHM of X, consistent with the direct measurement of the corresponding radiative lifetime, and demonstrates the superior operation of a single photon emitter based on biexciton emission. Recent experiments[22] have indicated an extended spin lifetime for excitons in quantum dots, which is longer than the radiative lifetime. Thus we can expect that the polarization states of the exciton and biexciton photon remain entangled after emission,[23] allowing a semiconductor source for entangled photon pairs to be developed.

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