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Photon antibunching at high temperature from a single InGaAs/GaAs quantum dot

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We report the observation of photon antibunching from a single, self-assembled InGaAs/GaAs quantum dot at temperatures up to 135 K. The second-order intensity correlation, $g^{(2)}(0)$, is measured to be less than 0.260 for temperatures up to 100 K. At 120 K, $g^{(2)}(0)$ increases to about 0.471, which is slightly less than the second-order intensity correlation expected from two independent single emitters. At 135 K, $g^{(2)}(0)$ is 0.667, which still indicates nonclassical light emission that is equivalent to having three independent single emitters. [DOI: 10.1063/1.1650032]

Emitters of single photons on demand are important for quantum key distribution (QKD) and low light level metrology. For practical implementation of QKD, it is especially important to have sources that operate at high temperatures. Several different approaches have recently demonstrated single photon emission, including epitaxial InGaAs/GaAs quantum dots (QDs),[1,2] epitaxial[3] and colloidal[4] CdSe quantum dots, GaAs interface fluctuation QDs,[5] single molecules,[6] and nitrogen vacancy (NV) color centers in artificial diamond.[7] Single molecules, colloidal CdSe QDs, and NV centers have all demonstrated room temperature single photon emission. However, both colloidal QDs and NV centers exhibit blinking,[4,7] which degrades the efficiency of the single photon source, and some single molecules also exhibit photobleaching, at which point the molecules no longer emit any photons. All of these methods are difficult to integrate with microcavities. Epitaxial InGaAs/GaAs QDs are attractive as single photon emitters for several reasons, including ease of fabrication and inclusion with monolithic microcavities, short spontaneous emission lifetimes, and the possibility of electrical injection.

To date, most of the studies of single photon emission from single InGaAs/GaAs QDs have been at low temperatures, typically less than 10 K. The use of microcavities to increase the photon capture efficiency means that the same QD cannot be investigated over a wide temperature range because of the differing temperature dependence of the cavity resonance and the QD bandgap. There are very few studies of high temperature (>50 K) single InGaAs/GaAs QD emission.[8] Recently, single epitaxial CdSe QDs demonstrated nonclassical light emission[3] up to 200 K, although the performance was substantially degraded above 40 K. In this letter we report our experimental results on single photon emission from single epitaxial InGaAs/GaAs QDs at temperatures ranging from 5.0 to 135.0 K. We demonstrate provable single emitter emission up to 120 K and photon antibunching up to 135 K, which is the highest temperature at which photon antibunching is reported for this system.

Our sample consists of a low areal density (about 1 $\mu\mathrm{m}^{-2}$) array of InGaAs QDs with $\mathrm{Al}_{0.2}\mathrm{Ga}_{0.8}\mathrm{As}$/GaAs barriers. Mesas of various sizes are wet etched on the sample to isolate various numbers of QDs. The mesas are imaged using a confocal microscope with an objective that has a numerical aperture of 0.40. Pulses from a mode-locked Ti:sapphire laser (850 nm, ~82 MHz, ~200 fs pulse width) are focused onto a mesa, and the light emitted from the QD is focused onto the input slits of a 0.3 m monochromator. A liquid nitrogen cooled charge coupled device (CCD) camera is used to record a photoluminescence (PL) spectrum. An internal mirror in the monochromator can be switched to direct the light emitted onto a Hanbury Brown–Twiss interferometer (HBTI) that consists of a 50/50 beamsplitter cube, two silicon photon counting avalanche photodiodes (APDs), and appropriate electronics. A slit on the output of the monochromator is used to adjust the spectral width of light incident on the HBTI to 1.2 nm (2.4 nm at the highest temperature). The start–stop time intervals from the APDs are recorded and binned with time resolution of 272 ps to form a histogram. The total efficiency of the system (defined here as the APD count rate divided by the laser pulse repetition rate) is around 10^{-4} because of the small fraction of emitted photons that are collected by the objective.

Figure 1 shows the temperature-dependent photoluminescence from a single QD captured by the CCD camera. Due to the above bandgap excitation of the GaAs, there are several emission peaks that are associated with excitons, charged excitons, and biexcitons from the QD. We will discuss the effect of these other peaks on the second-order intensity correlation below. Figure 1 shows the QD under the same excitation conditions that were used for measurements of second-order intensity correlation.

Figure 2 shows a histogram of counts made using the HBTI. We only show the peak at zero delay and a few of the adjacent peaks, but our measurement apparatus allows us to collect data with time separations up to 1115 ns, corresponding to about 90 peaks at a pulse repetition interval of 12 ns. The spontaneous emission lifetime of our QD is about 1 ns, so we can be almost certain that the QD is empty each time a pump pulse arrives. This histogram corresponds to the second-order intensity correlation under the condition of low collection efficiency.[9] The count rate of the APDs ranges from about 3.5×10^{3} counts/s at 50 K to about 1.1×10^{3} counts/s at 135 K. This includes the dark count rate of

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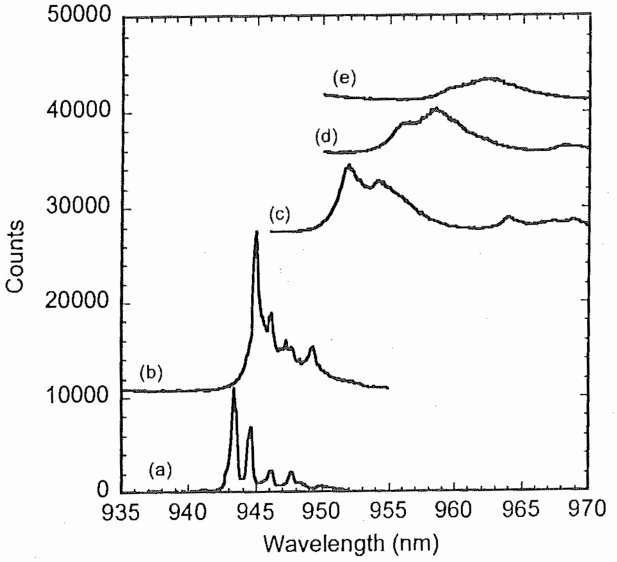

FIG. 1. Temperature-dependent optical spectra obtained from a 2×2 μm mesa using a CCD camera with 10 s integration time. These spectra are obtained under conditions identical to the photon correlation measurements shown in Fig. 2. The temperature and the average pump excitation intensity are (a) 5.0 K, 1.8 W/cm^2, (b) 50.0 K, 5.3 W/cm^2, (c) 100.0 K, 16.2 W/cm^2, (d) 120.0 K, 57.3 W/cm^2, and (e) 135.0 K, 84.3 W/cm^2. The spectra are vertically offset for clarity.

65 counts/s and 212 counts/s of the two APDs. Note that the count rate at 50 K is higher than at 5 K due to the fact that the QD is pumped harder at 50 K than at 5 K. When the same QD is excited with the same power density at 5 and 50 K, the count rate is about 25% higher at 5 K. However, this does lead to some degradation of the intensity correlation function as discussed below.

As shown in Fig. 2, the area of the peak at zero delay is much smaller than the area of all the other peaks. This is the signature that the photons are emitted one by one. The normalized peak area indicated in the caption of Fig. 2 is obtained by dividing the peak area at zero delay by the average area of all the other peaks (including the peaks not shown in Fig. 2). We also note that the peaks immediately adjacent to the peak at zero delay are somewhat larger than all of the other peaks, and these adjacent peaks also have a tail that does not go completely to zero before the next peak appears. The reason for these features is that the Si APDs will occasionally emit photons from the avalanche region after they detect a single photon.[10] Imperfect filtering and antireflection coatings on optics cause these emitted photons to be detected by one of the APDs.

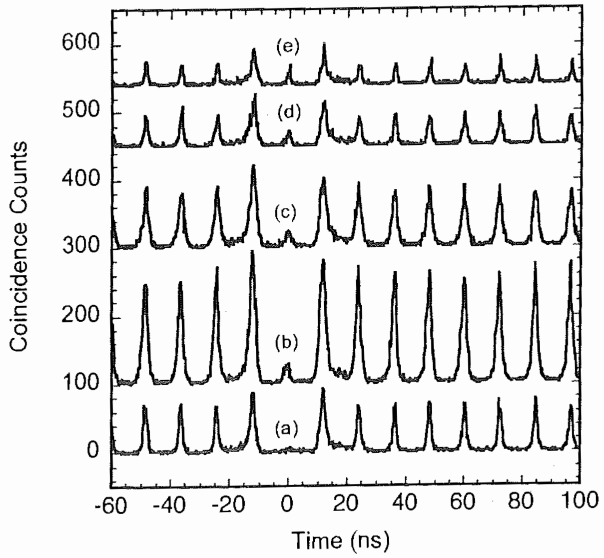

FIG. 2. Coincidence counts measured using a Hanbury Brown–Twiss interferometer. The temperatures and average pump laser intensity for each trace are those given in Fig. 1. The values of the second-order intensity correlation, $g^{(2)}(0)$, are (a) 0.087±0.009, (b) 0.177±0.013, (c) 0.260±0.024, (d) 0.471±0.067, and (e) 0.667±0.063. The uncertainty is primarily due to distribution of the peak areas used to compute the average peak area.

The best results are obtained at 5 K, in which $g^{(2)}(0)$ is measured to be 0.087±0.009. The second-order intensity correlation at zero delay increases gradually as the temperature increases and reaches a value of 0.260±0.024 at 100 K. At 120 K [Figs. 1(d) and 2(d)], the performance is reduced dramatically, with $g^{(2)}(0)$ increasing to 0.471±0.067. At 135 K [Figs. 1(e) and 2(e)], the single QD emission is quite weak. The average intensity of the Ti:sapphire laser has to be increased to about 84.3 W/cm^2 in order to obtain a count rate of about 1.1×10^3 counts/s on the APDs in the HBTI. At this temperature, the second-order intensity correlation increases to 0.667±0.063, which still indicates nonclassical light emission, but is no longer unambiguous proof of emission from a single quantum system.

The nonzero value of $g^{(2)}(0)$ is in part due to photons other than those that arise from the uncharged single exciton transition. These photons correspond to the peaks at longer wavelengths in the PL spectrum. We are able to accurately fit the spectra in Figs. 1(a)–1(c) to a sum of Lorentzians. The linear correlation coefficient, R, of our fit is greater than 0.95 for all three of these spectra. The fit allows us to determine the width and center wavelength of the peaks. By using the spectral width (1.2 nm) of the light incident on the HBTI and the fit values for the peaks, we are able to determine the fraction of counts in the spectra that are due to the uncharged single exciton peak, f_{ex}. This value is 0.960, 0.735, and 0.578 in Figs. 1(a)–1(c), respectively. The fraction of the area of the $g^{(2)}(0)$ peak that corresponds to emission of successive photons from the uncharged single exciton peak is simply $(f_{\mathrm{ex}})^2$. Therefore, if we were able to eliminate the contribution to the spectra due to the additional peaks, we would obtain $g^{(2)}(0)$ values of 0.080, 0.096, and 0.087 at quantum dot temperatures of 5, 50, and 100 K, respectively. This indicates the importance of reducing the biexciton emission from the quantum dot. We note here that we are unable to obtain a satisfactory fit to the spectra shown in Figs. 1(d) and 1(e) and were unable to perform the same analysis on these spectra. The uncharged single exciton emission peak is smaller than the additional peak at longer wavelengths in these two spectra, so it is likely that the value of f_{ex} is less than 0.50 for these two spectra. This sets an upper bound on the corrected $g^{(2)}(0)$ of 0.118 and 0.167 at quantum dot temperatures of 120 and 135 K, respectively.

These results show promise for high temperature operation of a single photon source based on InGaAs/GaAs QDs. To the best of our knowledge, there are no other reports of temperature-dependent photon antibunching measurements on single InGaAs/GaAs QDs. The highest reported temperature[11] is 44 K [$g^{(2)}(0)=0.38$ under continuous wave (cw) illumination]. The reason for this temperature is simply that the experiment was done on a QD in a cavity, and 44 K was the temperature at which the cavity resonance and the QD exciton emission peak were aligned. Single epitaxial CdSe QDs show photon antibunching up to 200 K (second-order intensity correlation of 0.81 at 200 K), but with poorer performance above 40 K. Our InGaAs QD shows an approximately linear increase in second-order intensity correlation with temperature up to 100 K, with a sharper increase above 100 K.

There are two reasons why the single InGaAs QD decreases in emission at higher temperatures, thereby requiring higher pump intensities and subsequently degrading the performance as a single photon source. First, the conduction band offset is relatively small, allowing thermionic emission of electrons out of the QDs into the InGaAs wetting layer. The QD that is reported in this letter was chosen in part for its separation from the wetting layer peak. Other QDs with shorter emission wavelengths have also been measured, but these QDs did not perform as well as the temperature increased. This suggests that carrier transfer to the wetting layer is a factor in the degradation of the second-order intensity correlation. Also, optical phonon scattering becomes more pronounced as the temperature increases. Using larger quantum dots with less quantum confinement energy or wider bandgap barriers can mitigate the conduction band offset problem. The phonon scattering problem could be mitigated by promoting faster radiative recombination times of the QD excitons by the use of a microcavity. Single electron-hole pair injection by electrical means[12] should improve the performance of an InGaAs QD single photon source by removing the possibility of charged exciton or biexciton formation.

In summary, we reported the measurement of photon antibunching from a single InGaAs QD up to 135 K and single emitter emission up to 120 K. This is a very high temperature for antibunched single photons from an InGaAs QD. These measurements indicate the potential for sources of single photons based on InGaAs QDs that operate at high temperature and show the importance of operating with a very low excitation level of the quantum dot to ensure that only one photon is emitted.

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[1] P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, L. D. Zhang, E. Hu, and A. Imamoglu, Science **290**, 2282 (2000).

[2] C. Santori, M. Pelton, G. Solomon, Y. Dale, and Y. Yamamoto, Phys. Rev. Lett. **86**, 1502 (2001).

[3] K. Sebald, P. Michler, T. Passow, D. Hommel, G. Bacher, and A. Forchel, Appl. Phys. Lett. **81**, 2920 (2002).

[4] P. Michler, A. Imamoglu, M. D. Mason, P. J. Carson, G. F. Strouse, and S. K. Buratto, Nature (London) **406**, 968 (2000).

[5] J. Hours, S. Varoutsis, M. Gallart, J. Bloch, I. Robert-Philip, A. Cavanna, I. Abram, F. Laruelle, and J. M. Gerard, Appl. Phys. Lett. **82**, 2206 (2003).

[6] B. Lounis and W. E. Moerner, Nature (London) **407**, 491 (2000).

[7] A. Beveratos, S. Kühn, R. Brouri, T. Gacoin, J.-P. Poizat, and P. Grangier, Eur. Phys. J. D **18**, 191 (2002).

[8] M. Bayer and A. Forchel, Phys. Rev. B **65**, 041308 (2002).

[9] J. G. Rarity, S. C. Kitson, and P. R. Tapster, in *Confined Photon Systems: Fundamentals and Applications*, edited by H. Benisty, J.-M. Gérard, R. Houdré, J. Rarity, and C. Weisbuch (Springer, Berlin, 1999), Vol. 531, p. 352.

[10] G. Ulu, A. V. Sergienko, and M. S. Ünlu, Opt. Lett. **25**, 758 (2000).

[11] A. Kiraz, P. Michler, C. Becher, B. Gayral, A. Imamoglu, L. D. Zhang, E. Hu, W. V. Schoenfeld, and P. M. Petroff, Appl. Phys. Lett. **78**, 3932 (2001).

[12] A. Imamoglu and Y. Yamamoto, Phys. Rev. Lett. **72**, 210 (1994).