

Photon antibunching from a single lithographically defined InGaAs/GaAs quantum dot

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We demonstrate photon antibunching from a single lithographically defined quantum dot fabricated by electron beam lithography, wet chemical etching, and overgrowth of the barrier layers by metalorganic chemical vapor deposition. Measurement of the second order autocorrelation function indicates $g^{(2)}(0) = 0.395 \pm 0.030$, below the 0.5 limit necessary for classification as a single photon source.

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Single photon emitters are important for quantum key distribution (QKD)^{1,2}, quantum metrology, and quantum information processing applications^{3,4}. Sources of single photons include epitaxial self-assembled quantum dots (QDs)⁵⁻⁷, colloidal quantum dots⁸, single molecules⁹, and nitrogen vacancy (NV) centers in diamond^{10,11}. Of these sources, epitaxial self-assembled QDs are attractive due to their ease of fabrication and incorporation within other photonic structures such as optical cavities. The random nature of the QD nucleation process, however, precludes the deterministic incorporation of a single QD within a cavity which is necessary to create an efficient single photon source or to perform quantum information processing operations at the single photon level. The self-assembly process also leads to variations in QD properties such as size, strain, and geometry which are correlated with electronic properties such as the biexciton binding energy, the fine structure splitting, and the emission wavelength^{12,13}.

Numerous techniques have been investigated for deterministically positioning a QD such as growth in etched nanoholes^{14,15}, atomic force microscopy¹⁶, and growth in inverted pyramidal recesses¹⁷. While these approaches have been shown to produce high optical quality site-controlled QDs, they are limited in scalability due to the bottom-up approach to fabrication – the QD properties depend critically on growth parameters and the properties of the prepatterned substrate such as the separation between QDs in the array and the individual pattern dimensions.

In this work, we present a novel top-down lithographic approach to the fabrication of single QDs which utilizes a combination of electron beam lithography, wet chemical etching of a preexisting quantum well, and regrowth of the barrier layers by metalorganic chemical vapor deposition (MOCVD). This top-down approach to QD fabrication

enhances the degree to which individual QD properties may be engineered and provides the site-control necessary for large-scale integration with other photonic structures.

The fabrication process begins with a base structure grown by molecular beam epitaxy (MBE) on an undoped (100) GaAs substrate and consists of a 20-period GaAs/AlAs distributed Bragg reflector stack, a 130 nm GaAs lower core, an 8 nm $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$ quantum well, and a 10 nm GaAs cap.

Electron beam lithography was performed with a scanning electron microscope with an acceleration voltage of 30 kV and beam current of 20 pA. PMMA was used as the electron beam resist. Regular arrays of dots were patterned in square lattices with various pitches ranging from 500 nm to 5 μm . The dot diameters were also varied by modifying the electron beam dose. After development, 20 nm of titanium metal was evaporated on the sample, followed by liftoff in acetone. The metal dot patterns were then transferred into the underlying quantum well layer using a phosphoric acid-based etchant. The etch was timed to provide an etch depth of 25 nm. After etching, the titanium was stripped in buffered hydrofluoric acid. Fig. 1 shows an array of 20 nm diameter QDs on a 500 nm pitch after stripping the etch mask. Following the etching step, the barrier layers were regrown in a low pressure MOCVD reactor and consist of a 130 nm GaAs upper core, an 80nm AlAs confinement layer, and a 10 nm GaAs cap.

Optical measurements were performed at a temperature of 4 K in a liquid helium cryostat. A modelocked Ti:sapphire laser (795 nm wavelength, 82 MHz repetition rate) was used as the excitation source. The pump beam was focused down to a spot size of approximately 4 μm with a 0.6 numerical aperture objective. Light emitted from the QD was collected by the same objective and focused onto the input slit of a 0.75 m

monochromator. An internal mirror in the monochromator was used to switch between a liquid nitrogen-cooled CCD camera for recording emission spectra, and a Hanbury-Brown-Twiss interferometer (HBTI) for measurement of the second order autocorrelation function. The HBTI consists of a 50/50 beamsplitter, two single photon avalanche diodes (SPADs), and timing electronics for recording the time interval between detection events on the two SPADs. The start-stop time intervals are recorded and binned with a resolution of 256 ps to form a histogram that is proportional to the second order intensity autocorrelation function.

Fig. 2 shows the emission spectrum of a low density QD array consisting of 35 nm diameter QDs on a 2.5 μm pitch at a pump power of 100 nW. Since the diameter of the pump spot is approximately 4 μm , we can be certain that we are primarily pumping a single QD in this array. The primary peak at 888.6 nm has a full width at half maximum of 0.16 nm or 260 μeV , significantly larger than the linewidths of typical self-assembled QDs¹⁸. While the origin of the larger linewidth requires further investigation, we suggest that it may be due to spectral diffusion caused by trapped charges near the etched interfaces of the QD¹⁹. As shown in the inset, the linear dependence of the integrated intensity on pump power below 200 nW suggests that this spectral line is due to emission from a single exciton state. The smaller peaks on the long wavelength side of the primary peak are likely due to emission from adjacent QDs in the array which are at the edge of the pump beam. This was verified by moving the sample stage and observing the simultaneous decay of the primary peak and increase in the intensity of the adjacent peaks. In addition, the intensity of the adjacent peaks is very weakly correlated with

pump power in comparison to the single exciton line, making it unlikely that these peaks correspond to emission from biexciton or charged exciton states from the same QD.

Fig. 3 shows the second order autocorrelation function measured on the single exciton line at 888.6 nm in Fig. 2. The measurement was performed at a pump power of 175 nW. The entrance and exit slits of the monochromator were set to pass a spectral bandwidth of ~ 0.1 nm to the HBTI. The count rate on each SPAD was approximately 700 Hz for the duration of the measurement. Although the timing electronics allow us to collect data for time separations τ up to 16 μ s, we show only the first few peaks around $\tau = 0$ for clarity. The value of $g^{(2)}(0)$ can be computed by taking the area of the peak at zero time delay and dividing by the average area of all other peaks. Performing this calculation without subtracting the background due to dark counts on the SPADs yields $g^{(2)}(0) = 0.395 \pm 0.030$. This value is below the 0.5 limit necessary for classification as a single photon source²⁰. If background subtraction is performed, we obtain $g^{(2)}(0) = 0.314 \pm 0.029$. The remaining counts at zero time delay may be due to the weak background emission around the single exciton line as shown in Fig. 2, as well as imperfect spectral filtering of the two longer wavelength peaks caused by emission from adjacent QDs in the array.

In summary, we have demonstrated the first observation of single photon emission from a novel type of lithographically defined quantum dot. The fabrication process allows for control of the geometry, emission wavelength, and position of the QD. The MOCVD regrowth process should allow such a QD to be easily incorporated into photonic structures such as optical microcavities. The ability to fabricate multiple QDs having nearly identical properties may also prove useful for generating quantum states of light that are not possible with self-assembled QDs due to statistical variations, such as an

on-demand source of two or more entangled photons using two or more identical QDs. In the future it may also provide a scalable approach to performing on-chip quantum information processing.

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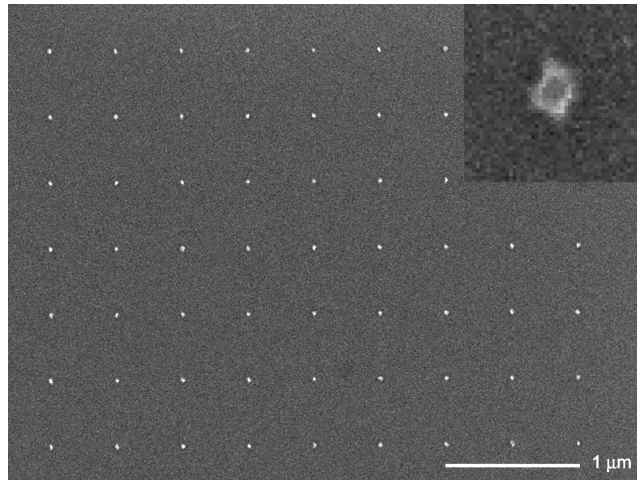


Fig. 1 An array of 20 nm diameter etched QDs on a pitch of 500 nm. The inset shows a magnified image of a single QD.

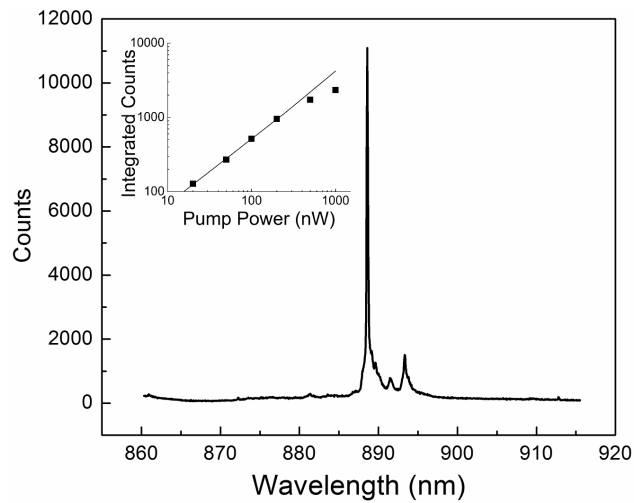


Fig. 2 Emission spectrum of a 35 nm QD in a 2.5 μm pitch array. The inset shows the integrated intensity of the exciton line at 888.6 nm as a function of pump power. The solid line is a linear fit to the data below 200nW.

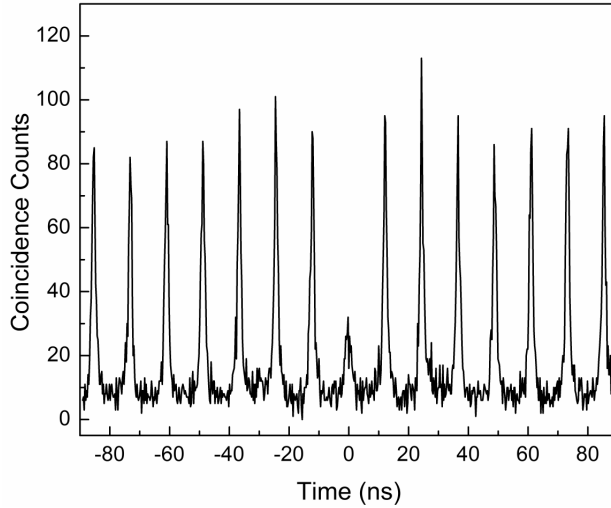


Fig. 3 Second order autocorrelation function $g^{(2)}(\tau)$ measured on the exciton line at 888.6 nm in the emission spectrum of Fig. 2.

REFERENCES

- ¹N. Gisin, G. Ribordy, W. Tittel, and H. Zbinden, *Rev. Mod. Phys.* **74**, 145 (2002).
- ²E. Waks, K. Inoue, C. Santori, D. Fattal, J. Vuckovic, G. S. Solomon, and Y. Yamamoto, *Nature* **420**, 762 (2002).
- ³A. Imamoğlu, *Physica E: Low-dimensional Systems and Nanostructures* **16**, 47 (2003).
- ⁴A. Kiraz, A. Atatüre, and A. Imamoğlu, *Phys. Rev. A* **69**, 032305 (2004).
- ⁵Richard P. Mirin, *Appl. Phys. Lett.* **84**, 1260 (2003).
- ⁶C. Santori, D. Fattal, J. Vuckovic, G. S. Solomon, and Y. Yamamoto, *New J. Phys.* **6**, 89 (2004).
- ⁷P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, L. Zhang, E. Hu, and A. Imamoğlu, *Science* **22**, 2282 (2000).
- ⁸X. Brokmann, G. Messin, P. Desbiolles, E. Giacobino, M. Dahan, and J. P. Hermier, *New J. Phys.* **6**, 99 (2004).

- ⁹B. Lounis and W. E. Moerner, *Nature* **407**, 491 (2000).
- ¹⁰C. Kurtsiefer, S. Mayer, P. Zarda, and H. Weinfurter, *Phys. Rev. Lett.* **85**, 290 (2000).
- ¹¹T. Babinec, B. J. M. Hausmann, M. Khan, Y. Zhang, J. R. Maze, P. R. Hemmer, and M. Lončar, *Nature Nanotech.* **5**, 195 (2010).
- ¹²R. J. Young, R. M. Stevenson, A. J. Shields, P. Atkinson, K. Cooper, D. A. Ritchie, K. M. Groom, A. I. Tartakovskii, and M. S. Skolnick, *Phys. Rev. B* **72**, 113305 (2005).
- ¹³D. Sarkar, H. P. van der Meulen, J. M. Calleja, J. M. Becker, and R. J. Haug, *J. Appl. Phys.* **100**, 023109 (2006).
- ¹⁴C. Schneider, A. Huggenberger, T. Sünner, T. Heindel, M. Strauß, S. Göpfert, P. Weinmann, S. Reitzenstein, L. Worschech, M. Kamp, S. Höfling, and A. Forchel, *Nanotechnology* **20**, 434012 (2009).
- ¹⁵P. Atkinson, M. B. Ward, S. P. Bremner, D. Anderson, T. Farrow, G. A. C. Jones, A. J. Shields, and D. A. Ritchie, *Jpn. J. Appl. Phys.* **45**, 2519 (2006).
- ¹⁶H. Z. Song, T. Usuki, T. Ohshima, Y. Sakuma, M. Kawabe, Y. Okada, K. Takemoto, T. Miyazawa, S. Hirose, Y. Nakata, M. Takatsu, and N. Yokoyama, *Nanoscale Res. Lett* **1**, 160 (2006).
- ¹⁷A. Mohan, M. Felici, P. Gallo, B. Dwir, A. Rudra, J. Faist, and E. Kapon, *Nature Photonics* **4**, 302 (2010).
- ¹⁸K. Leosson, J. R. Jensen, J. M. Hvam, and W. Langbein, *Phys. Stat. Sol. (b)* **221**, 49 (2000).
- ¹⁹J. Seufert, R. Wigand, G. Bacher, T. Kummell, A. Forchel, K. Leonardi, and D. Hommel, *Appl. Phys. Lett.* **76**, 1872 (2000).
- ²⁰R. Loudon, *The Quantum Theory of Light* (Oxford University Press, New York, 1983).