

Registration of single quantum dots using cryogenic laser photolithography

Kwan H. Lee, Alex M. Green, Robert A. Taylor,^{a)} David N. Sharp, Jan Scrimgeour, Olivia M. Roche, Jong H. Na, Anas F. Jarjour, and Andrew J. Turberfield
Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom

Frederic S. F. Brossard and David A. Williams
Hitachi Cambridge Laboratory, Hitachi Europe Ltd., Madingley Road, Cambridge CB3 0HE, United Kingdom

G. Andrew D. Briggs
Department of Materials, University of Oxford, Parks Road, Oxford OX1 3PH, United Kingdom

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We have registered the position of single InGaAs quantum dots using a cryogenic laser photolithography technique. This is an important advance towards the reproducible fabrication of solid-state cavity quantum electrodynamic devices, a key requirement for commercial exploitation of quantum information processing. The quantum dot positions were registered with an estimated accuracy of 50 nm by fabricating metal alignment markers around them. Photoluminescence spectra from quantum dots before and after marker fabrication were identical except for a small redshift (~ 1 nm), probably introduced during the reactive ion etching. © 2006 American Institute of Physics. [DOI: 10.1063/1.2202193]

Solid state systems such as excitons or spin states in quantum dots (QDs) have attracted much attention as potential qubits.¹ A common feature in many of these schemes has been the requirement for the generation of single photons on demand² with many experiments exploiting cavity quantum electrodynamic (CQED) solid state devices consisting of QDs positioned in optically structured microcavities.^{3–5} A key requirement to achieving effective coupling between the cavity mode and the QD exciton is the accurate alignment; the electric field antinode of the cavity must be located at or very near the QD (~ 100 nm). To date, most solid state CQED devices have relied on chance to achieve alignment between randomly positioned QDs and the cavity. For example, Yoshie *et al.*⁴ fabricated $\sim 30\,000$ photonic crystal nanocavities on a high density QD sample ($\sim 3.5 \times 10^{10} \text{ cm}^{-2}$). The low yields ($<1\%$) inherent in the chance method, and the time taken to find successful devices, make it unsuitable for commercial exploitation. Furthermore, the use of high QD densities reduces the yield of true single-photon emitters, as nearby QDs may also couple to the cavity mode. It is desirable to develop a technique that enables the fabrication of an aligned cavity by marking, or registering, the spatial position of a QD.^{6,7}

In this letter we present an innovative technique for the registration of a single QD, located by low-temperature spectroscopy, by the creation of alignment markers by photolithography at 4 K. Markers are created by two-photon absorption (TPA) laser photolithography of SU-8, a negative photoresist widely used in the microelectromechanical systems community.⁸ TPA photolithography exploits nonlinear absorption by SU-8 of high-peak-power laser pulses at a wavelength that is too long to expose the resist by single-photon absorption.⁹ The possibility of separating the excitation of the photoacid-generator photoinitiator, which can occur at low temperature, and acid-catalyzed polymerization, which can be delayed and initiated at a higher temperature without loss of resolution, make SU-8 an attractive candidate

for low-temperature exposure.¹⁰ Advantages of this technique are that it does not rely on chance, is compatible with low-density QD samples, and allows selection of QDs with desired properties (e.g., wavelength, lifetime). We discuss the details of the QD registration process and investigate its accuracy.

The apparatus used for microphotoluminescence (μPL) and TPA laser photolithography is illustrated in Fig. 1. The sample was mounted in a continuous-flow liquid helium microscope cryostat (Janis ST-500), which was mounted on a micrometer XY stage. The primary light source for both spectroscopy and lithography was an 80 MHz, 800 nm

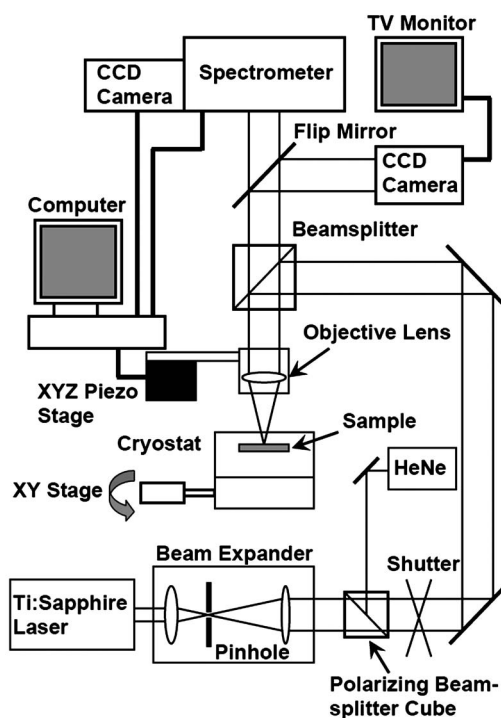


FIG. 1. Schematic diagram of the apparatus used to perform microphotoluminescence spectroscopy and laser photolithography.

^{a)}Electronic mail: r.taylor@physics.ox.ac.uk

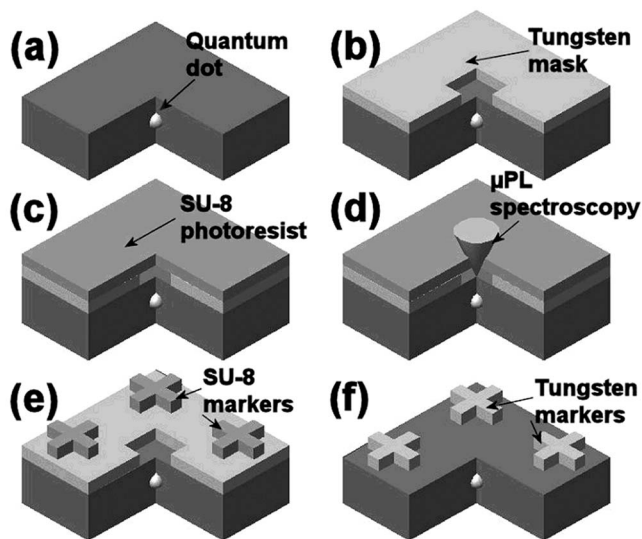


FIG. 2. Schematic diagram of the QD registration process. A QD sample (a) is coated with a tungsten mask (b) and then SU-8 photoresist is spin-coated on (c). μ PL locates the QD (d) and an alignment marker is patterned in the SU-8 (e). Subsequent RIE forms the final tungsten marker (f).

mode-locked Ti:sapphire laser with ~ 100 fs pulse width (Spectra Physics Mai-Tai). To achieve optimum resolution the beam was passed through a $75\text{ }\mu\text{m}$ diameter pinhole and expanded by a factor of ~ 1.5 . The laser pulses were focused by a 0.5 numerical aperture, $\times 100$ objective lens (Mitutoyo), which was mounted on a piezoelectric XYZ stage with $100\text{ }\mu\text{m}$ travel along each axis and 1 nm resolution (PI P-611.S). Drift of the sample position relative to the focus of the microscope was measured to be $\sim 1.5\text{ nm/min}$. Light collected through the objective was dispersed by a 0.3 m spectrograph, equipped with a 1200 grooves/mm grating, to give a spectral resolution of $\sim 0.7\text{ meV}$. PL was detected by a Peltier-cooled charge-coupled device (CCD) (Andor DU-420). μ PL and TPA laser photolithography were performed using the same excitation source: for TPA the beam power was $\sim 20\text{ mW}$ ($\sim 0.2\text{ nJ/pulse}$) at the focus and each voxel was exposed for $\sim 10\text{ ms}$; for μ PL the beam was attenuated by a factor of 10^5 . A secondary light source, a 633 nm continuous-wave He-Ne laser, was used for tests of registration accuracy as it could be focused to a smaller spot.

The registration process consists of four steps: (1) preparation of a metal layer with clear apertures on the sample; (2) μ PL spectroscopy to locate a QD within an aperture; (3) laser exposure of a resist layer to define alignment markers in the surrounding metal; and (4) fabrication of the alignment markers. Figure 2 illustrates this process. A metallic mask was formed by depositing 60 nm of tungsten on a low-density ($\sim 10^8\text{ cm}^{-2}$) InGaAs QD sample in a regular grid consisting of $15 \times 15\text{ }\mu\text{m}^2$ apertures separated by $100\text{ }\mu\text{m}$, using conventional photolithography [Figs. 2(a) and 2(b)]. Tungsten was chosen as it could be removed using reactive ion etching (RIE) and because its high atomic number will ensure good visibility of alignment markers in an electron beam writer used in subsequent cavity fabrication. The sample was then spin-coated with a uniform $2\text{ }\mu\text{m}$ layer of SU-8 and prebaked at $95\text{ }^\circ\text{C}$ for 2 min [Fig. 2(c)].

The positions and spectral characteristics of QDs were measured using μ PL [Fig. 2(d)]. QDs were located by measuring the spatial variation of the PL intensity at the exciton peak while scanning the microscope objective. Figure 3(a)

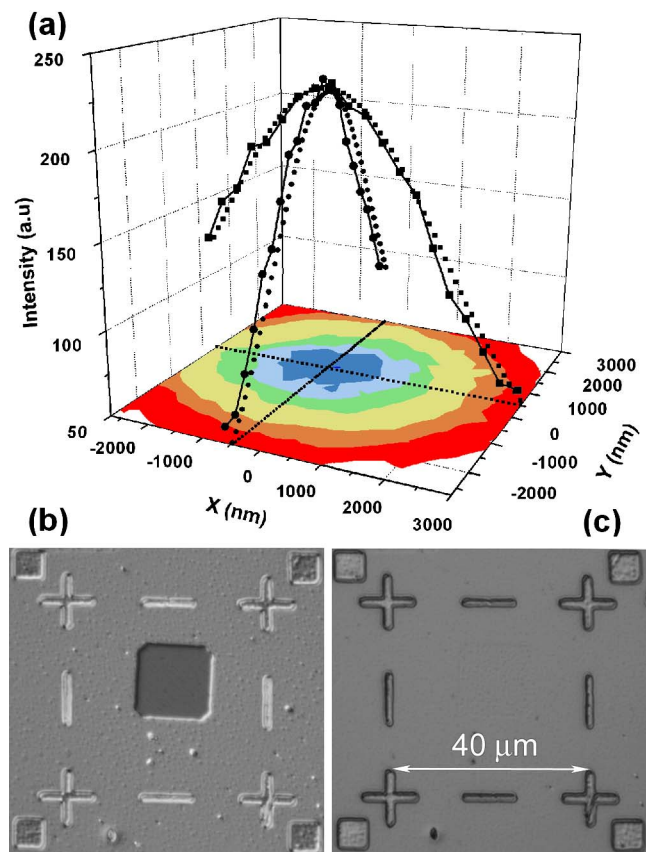


FIG. 3. (Color online) (a) Spatial intensity map of a QD exciton using 800 nm excitation. The spectral integration was over a $\sim 0.3\text{ nm}$ range centered at 875 nm [see Fig. 4(a)] and the position step was $0.3\text{ }\mu\text{m}$. Also, optical microscope images of the registration pattern before (b) and after (c) reactive-ion etching showing the pattern in SU-8 and tungsten, respectively.

shows a map of the intensity of PL from a single QD, which was obtained by scanning the focused laser spot over a $6 \times 6\text{ }\mu\text{m}^2$ area (detection bandpass: 0.3 nm centered at 875 nm , step size: 300 nm). The width of this feature is largely determined by the width of the Gaussian laser spot, which is much broader than the dimensions of the QD. Figure 3(a) shows orthogonal sections through the intensity distribution and the results of fitting to a Gaussian. The center of the intensity peak was located to within $\pm 25\text{ nm}$ using standard procedures for the localization of isolated fluorescent probes.¹¹ We have performed repeated measurements (eight times) on a single QD and have found that the R^2 varied from 0.990 to 0.924 with the peak uncertainty varying between ± 16 and $\pm 35\text{ nm}$. The uncertainty in the QD center position combines the uncertainty in fitting the center of the PL distribution with the effects of $\sim 15\text{ nm}$ drift of the apparatus during the 10 min acquisition time for the PL measurement. We estimate the QD registration accuracy to be better than $\pm 50\text{ nm}$.

After the location of the QD center, the beam power was increased and the alignment markers were patterned by moving the piezoelectric stage—on which the microscope objective was mounted—using a computer program written in LABVIEW. The exposed samples were postbaked at $95\text{ }^\circ\text{C}$ for 5 min , developed in propylene glycol methyl ether acetate, rinsed in isopropanol, and blown dry. Figure 3(b) is an optical micrograph showing the developed SU-8 alignment markers. The alignment pattern consisted of cross, line, and square shapes, with a minimum feature size of $\sim 1\text{ }\mu\text{m}$. The

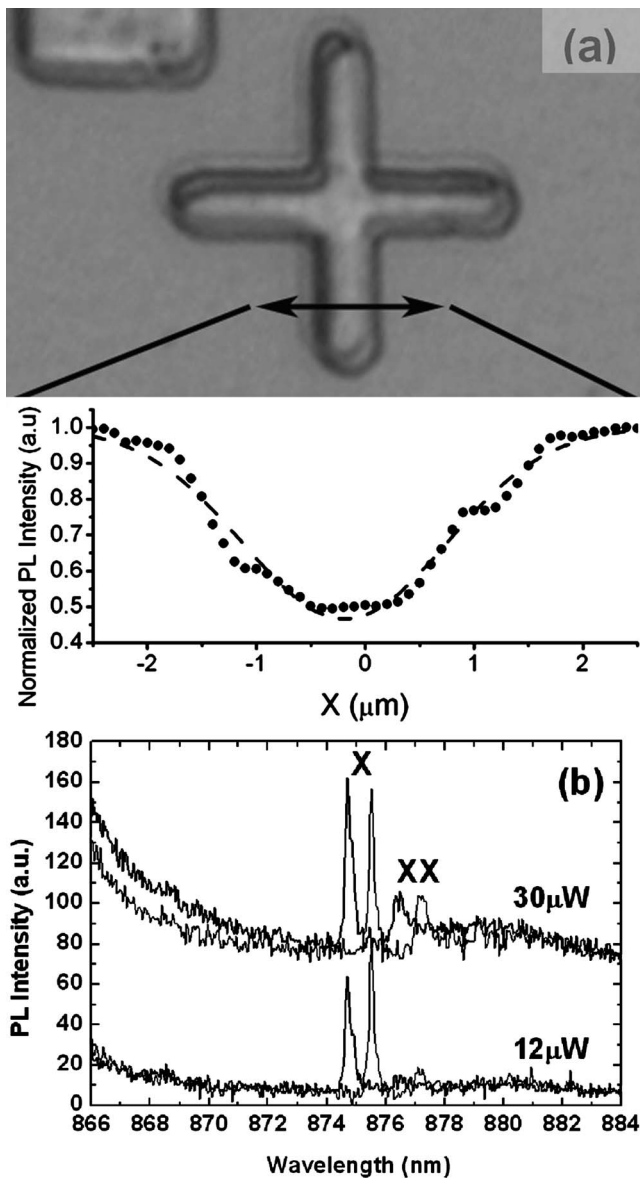


FIG. 4. (a) Scan across one of the cross arms and the corresponding intensity profile, with the fitted curve shown as a dashed line. (b) QD spectra taken before (black line) and after (gray line) registration marker fabrication, with the spectra obtained at two excitation powers and the biexciton (XX) peak clearly visible.

crosses (separated by $40\text{ }\mu\text{m}$) and lines were designed to enable the accurate relocation of the pattern center, so these were written initially (requiring 2 min) to minimize any errors due to apparatus drift during the writing process. The square outside each cross was designed to increase the visibility of the marker and to indicate its orientation. The alignment pattern was transferred into the tungsten layer by RIE using a $\text{CF}_4:\text{O}_2$ 10:1 gas mixture for 1 min. Any remaining SU-8 was removed using oxygen plasma ashing. The resulting tungsten alignment markers are shown in Fig. 3(c).

In future work electron beam lithography will be used for the fabrication of photonic crystal microcavities around the chosen QD. To complete this proof-of-concept study, we have used the apparatus shown in Fig. 1 to check the accuracy of the registration technique. The sample was cooled to 4 K and the QD was relocated by means of the alignment markers. The piezoelectric stage was used to scan the focus of the microscope objective over the arms of the crosses while recording the PL signal from the InAs wetting layer. A

$0.3\times 5\text{ }\mu\text{m}^2$ scan of a cross with 100 nm step size took ~ 3 min. By fitting multiple cross sections through all four arms [Fig. 4(a)] it was possible to locate the center of a cross with an uncertainty of ± 30 nm. Drift during these measurements was partially compensated by remeasuring the position of the QD between each scan of an alignment cross. The experimental error in determining the center of the alignment pattern by this technique was estimated to be ± 150 nm and the measured distance between this point and the observed position of the QD was consistent with this estimate, indicating that the fabrication tolerance is below the limit of resolution of this measurement.

Figure 4(b) shows the spectra from a typical registered QD before and after the marker fabrication process. A processing-induced redshift varying between 0.5 and 1.4 nm was observed from QDs across the whole sample and from different samples. A likely cause of the redshift is a Stark shift due to electric fields set up by the creation of surface states during RIE.¹² The second peak in Fig. 4(b) is due to the biexciton (XX) transition; this was verified by measuring the PL intensity dependence with excitation power.

In summary, we have demonstrated the registration (estimated ± 50 nm accuracy) and reacquisition (estimated ± 150 nm accuracy) of low-density InGaAs QDs using a laser cryogenic-photolithography technique. Our technique involved using a computer controlled piezoelectric stage to find QDs and then write alignment patterns via two-photon exposure of the SU-8 photoresist at a temperature of 4 K. This technique is transferable to other sample types and material systems.

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¹G. Ortner, M. Bayer, Y. Lyanda-Geller, T. L. Reinecke, A. Kress, J. P. Reithmaier, and A. Forchel, *Phys. Rev. Lett.* **94**, 157401 (2005).

²A. Kiraz, M. Atatüre, and A. Imamoglu, *Phys. Rev. A* **69**, 032305 (2004).

³A. Imamoglu, D. D. Awschalom, G. Burkard, D. P. DiVincenzo, D. Loss, M. Sherwin, and A. Small, *Phys. Rev. Lett.* **83**, 4204 (1999).

⁴T. Yoshie, A. Scherer, J. Hendrickson, G. Khitrova, H. M. Gibbs, G. Rupper, C. Ell, O. B. Shchekin, and D. G. Deppe, *Nature (London)* **432**, 200 (2004).

⁵J. P. Reithmaier, G. Sek, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. V. Keldysh, V. D. Kulakovskii, T. L. Reinecke, and A. Forchel, *Nature (London)* **432**, 197 (2004).

⁶A. Badolato, K. Hennessy, M. Atatüre, J. Dreiser, E. Hu, P. M. Petroff, and A. Imamoglu, *Science* **308**, 1158 (2005).

⁷A. Kiraz, C. Reese, B. Gayral, L. Zhang, W. V. Schoenfeld, B. D. Gerardot, P. M. Petroff, E. L. Hu, and A. Imamoglu, *J. Opt. B: Quantum Semiclassical Opt.* **5**, 129 (2003).

⁸K. Y. Lee, N. LaBianca, S. A. Rishton, S. Zolgharnain, J. D. Gelorme, J. Shaw, and T. H.-P. Chang, *J. Vac. Sci. Technol. B* **13**, 3012 (1995).

⁹W. H. Teh, U. Dürig, G. Salis, R. Harbers, U. Drechsler, R. F. Mahrt, C. G. Smith, and H.-J. Güntherodt, *Appl. Phys. Lett.* **84**, 4095 (2004).

¹⁰K. H. Lee, A. M. Green, R. A. Taylor, D. N. Sharp, A. J. Turberfield, F. S. F. Brossard, D. A. Williams, and G. A. D. Briggs, *Appl. Phys. Lett.* **88**, 143123 (2006).

¹¹R. E. Thompson, D. R. Larson, and W. W. Webb, *Biophys. J.* **82**, 2775 (2002).

¹²B. C. Qiu, B. S. Ooi, A. C. Bryce, S. E. Hicks, C. D. W. Wilkinson, R. M. De La Rue, and J. H. Marsh, *International Conference on Indium Phosphide and Related Materials*, Cape Cod (IEEE, Piscataway, NJ, 1997), p. 578.