Single electron-spin memory with a semiconductor quantum dot

Robert J Young\textsuperscript{1,3}, Samuel J Dewhurst\textsuperscript{1,2}, R Mark Stevenson\textsuperscript{1}, Paola Atkinson\textsuperscript{2}, Anthony J Bennett\textsuperscript{1}, Martin B Ward\textsuperscript{1}, Ken Cooper\textsuperscript{2}, David A Ritchie\textsuperscript{2} and Andrew J Shields\textsuperscript{1}

\textsuperscript{1} Toshiba Research Europe Limited, 260 Science Park, Cambridge CB4 0WE, UK
\textsuperscript{2} Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, UK
E-mail: robert.young@crl.toshiba.co.uk

New Journal of Physics 9 (2007) 365
Received 15 June 2007
Published 9 October 2007
Online at http://www.njp.org/
doi:10.1088/1367-2630/9/10/365

Abstract. We show storage of the circular polarization of an optical field, transferring it to the spin-state of an individual electron confined in a single semiconductor quantum dot. The state is subsequently read out through the electronically-triggered emission of a single photon. The emitted photon shares the same polarization as the initial pulse but has a different energy, making the transfer of quantum information between different physical systems possible. With an applied magnetic field of 2 T, spin memory is preserved for at least 1000 times more than the exciton’s radiative lifetime.

Efficient transfer of qubits is a necessity of any useful quantum information application and requires a powerful link between stored qubits and transmitted quantum data. Photons are an ideal choice for the ‘flying qubits’ required in applications such as quantum cryptography [1] and distributed quantum computing [2]. Quantum information processing, however, requires qubit operations and storage. Multiple qubit operations are non-trivial with photons as photon–photon interactions are difficult to achieve in practice. Linear optics quantum computing [3] does address this, though photonic memory is still a limiting factor. Conversely atomic and solid state spins [4] are relatively easy to manipulate and store but difficult to transfer over long distances. Storing excitons excited by single photons in semiconductor quantum dots [5] could provide a good interface between flying and stationary qubits. Additionally, the semiconductor nature of quantum dots facilitates integration into complex structures which are both compact and robust [6]–[8], expanding their appeal as an interface between optical
Figure 1. Simple schematic diagrams illustrating how the conduction (C) and valence (V) bands around a quantum dot can be manipulated to store single electrons. Writing mode: a single dot is quasi-resonantly excited with a circularly polarized weak pulse of light, the device is biased to remove holes (open circles) from the dot via tunnelling, the polarization of the pulse is stored on the spin-state of the confined electron (filled circles). Readout mode: The device is biased to return a pair of holes to the quantum dot, radiative recombination results in the emission of a single photon whose polarization is dictated only by the electron’s spin-state. As an example, right-hand circularly polarized (R) incident photons are labelled, black arrows illustrate spin orientations.

and solid-state quantum information. To date a wide range of quantum dot devices have been developed; diode structures to provide electrically triggered single photon emission [7], optical cavities enabling strong coupling between the photonic and excitonic modes [9]–[11], resonant tunnelling diodes allowing single photon counting [12] and devices capable of emitting polarization-entangled photon pairs [13]–[15].

The optical memory we have developed employs a single quantum dot for its active element and is operated as illustrated in figure 1. A single quantum dot is embedded in the intrinsic region of a diode. This is biased to make the tunnelling rate of heavy-holes from the quantum dot dominate over the radiative recombination rate of exciton states confined by the dot. A tunnel barrier on the negative side of the diode inhibits electrons from leaving the dot. A weak laser pulse can therefore be used to populate a single electron with a pure spin state into the quantum dot. This is stored until a pair of heavy-holes is returned by the application of an ac voltage pulse. Radiative decay of the positively charged exciton follows, leading to the emission of a single photon whose polarization is determined by the spin-state of the single electron.

Figure 2(a) illustrates the device design we used. Molecular beam epitaxy was used to grow a low density (<1 µm⁻²) layer of InAs quantum dots embedded in GaAs [5]. A 1λ planar optical cavity [16] designed with a mode at ~900 nm was formed by 12 repeats of alternating quarter-wavelength thick layers of GaAs/AlAs distributed Bragg reflectors below the dot layer and two repeats of the same mirror structure above the dot layer. The top mirror was p-doped with carbon and the two repeats of the bottom mirror were n-doped with silicon. Electrical contacts to the doped regions allow a field to be applied perpendicularly to the growth plane
Figure 2. (a) An illustration of the sample design. A low-density layer of quantum dots (QDs) is embedded in an optical cavity formed by n- and p-doped distributed Bragg reflectors (DBRs). Apertures in an aluminium mask allow access to single quantum dots, top (TC) and bottom (BC) electrical contacts are labelled. (b) Photoluminescence spectra taken from a single quantum dot as the bias across the dot is swept. Blue (white) areas indicate high (low) emission intensity. Emission from single positively (negatively) charged $X^+$ ($X^-$) excitons and the neutral exciton $X$ and biexciton $X_2$ are identified. The Zeeman interaction induces an energy separation between states of opposing spin, this is clearly visible for each of the labelled exciton states.

of the quantum dots. The dot layer was situated on top of a 5 nm layer of GaAs above an Al$_{0.98}$Ga$_{0.02}$As/Al$_{0.5}$Ga$_{0.5}$As superlattice; forming a tunnel barrier, restricting electrons from reaching the n-doped region. An aluminium shadow mask on top of the sample contains an array of $\sim 2 \mu m$ diameter circular apertures; these allowed single quantum dots to be repeatedly accessed.

The sample was cooled to $< 10 K$ in a continuous flow helium-4 cryostat and excited optically with weak $\sim 100 \text{ ps}$ pulses from a multimode diode laser emitting at $869 \pm 5 \text{ nm}$. A microscope objective was used to both focus the excitation laser to a $\sim 1 \mu m^2$ spot on the surface of the device and collect emitted photoluminescence. The photoluminescence was guided into a spectrometer and to a nitrogen-cooled charge-coupled device for wavelength-resolved spectroscopy.

Figure 2(b) shows photoluminescence collected from a single quantum dot in the device as a function of the dc bias applied to the diode. Emission from the neutral exciton ($X$, an
Figure 3. (a) Time-resolved emission measured directly from the laser and photoluminescence from the positively charged exciton in a single quantum dot with an applied dc bias. (b) Time-resolved emission from the same dot with a $-0.05$ V dc bias to remove holes from the quantum dot. 0.2 to 1 $\mu$s (as labelled) after the laser pulse an ac pulse was applied to inject a pair of holes to the dot. Emission from the $X^+$ state is now found to be delayed and in-phase with the ac pulse.

electron–hole pair) and biexciton ($X_2$, two electron-hole pairs) states is maximized with a dc bias of $\sim 1.25$ V. The dominant charge state can be switched by adjusting the voltage by 0.15 V below (above) this bias in which case emission is predominantly found to be from the negatively (positively) charged exciton $X^-(X^+)$ state. If a bias is applied which is significantly outside this range then no emission is measured. There is evidence of emission from exciton states containing more carriers, such as the lines visible at $\sim 898$ nm, though these are generally weak. The application of a 2 T magnetic field perpendicular to the dot layer introduces a Zeeman interaction between states of opposing angular momentum orientation, separating them in energy [17]. This results in the emission of all four of the exciton states, $X^-, X, X_2$ and $X^+$ splitting into well resolved doublets. The identification of the initial states responsible for the photoluminescence measured from small InAs quantum dots similar to those studied here has been the focus of other studies [18].

To measure time-resolved emission from the $X^+$ state, it was selected with a spectrometer and directed into an avalanche photodiode (APD). A single photon counting module measured the time delay between the electronic output of the APD and a trigger pulse from the laser to build a histogram of photon arrival times.

Single electron storage and readout with a quantum dot is demonstrated in figure 3. The black line in figure 3(a) shows time resolved emission from the $X^+$ state of a single quantum dot. A dc bias was applied so that emission from this state was triggered by the laser pulse which was set at a repetition rate of 80 MHz. The red trace measures the transient due to the $\sim 100$ ps laser pulse scattered from the sample’s surface. The resolution of this decay curve is limited by the jitter time of the detector used.
Figure 3(b) shows emission collected from the same exciton state and dot. The frequency of the laser pulses was lowered to 1 MHz and an ac voltage pulse was applied on top of a small negative dc bias. A pulse-pattern generator was used to both provide a clock signal for the laser used to excite the dot and for the ac pulses applied to the sample. The delay between the laser write pulse and the ac read pulse was varied from 0.2 to 1 µs. The delay of the observed luminescence correlates with the delay of the ac pulse, thus emission is clearly triggered by the readout pulse and not the laser light. The variation in the area of the readout peaks in the traces shown in figure 3(b) is a result of fluctuations in the intensity of the write laser pulse, a result of small lateral movements of the sample in our cryostat.

To confirm that the measured signal can indeed be attributed to electron storage by the quantum dot and is not directly induced electroluminescence by the ac pulse, the laser light was removed and no signal was measured (results not shown). Fine control of the applied dc and ac biases allowed the emission from either the neutral or the positively charged exciton states to be delayed. The latter was chosen for readout as it prevents the formation of long-lived dark states, which would severely limit the speed at which readout can be performed. Limitations in the circuit used to deliver ac pulses, caused by impedance mismatch between the circuitry and the device, meant the pulse was attenuated somewhat before reaching the device. A larger ac pulse was required to put the device into the charge state of a given dc bias.

The polarization of the delayed emission from the X\textsuperscript{+} state was measured under different pump polarization conditions. To excite the sample with right- (left-) hand circularly polarized light the linearly polarized laser was passed through a broadband quarter-waveplate placed directly before the microscope objective orientated with its fast axis at +45° (−45°), degrees relative to the laser polarization. The quarter-waveplate also served to rotate circularly polarized emission from the sample into a linear basis. A half-waveplate and linear polarizer were placed directly before the spectrometer, the angle of the latter was fixed to maximize transmission through the spectrometer. Two measurements were taken with the half-waveplate at 0 and +45°; to analyse the emission in independent cross-polarized bases.

With unpolarized excitation each pair of lines shown in figure 2(b) are found, as expected, to be polarized in opposite circular polarizations. When the polarization of the excitation laser is linear, equal numbers of up- and down-spin electrons are excited in the device and the spin of the electron confined by the quantum dot is random. As a result of this the delayed emission from the two pure-spin components of the X\textsuperscript{+} state are equal. Figure 4(a) demonstrates this, showing equal photoluminescence from both spin states after ∼1 µs delay following excitation with a horizontally (H) polarized pump. This result is in stark contrast to those shown in figure 4(b) and (c) obtained using a circularly polarized pump, which excites spin-polarized electrons. Here one of the two components of the positively charged exciton is found to be much more intense than the other depending on the orientation of the pump polarization. The polarization of the emission correlates with the pump polarization. These measurements demonstrate that the electron spin is well preserved during the process of relaxing into the quantum dot following non-resonant excitation, removal of the hole from the dot and subsequent re-injection of a pair of holes.

The degree of polarization memory is defined as \(|(I_L - I_R) / (I_L + I_R)|\), where \(I_L (I_R)\) is the area of the left-hand (right-hand) circularly polarized X\textsuperscript{+} emission peak. This was found to be 80 ± 10% for both circular pump polarizations and constant within the delay times, 0.2–1 µs, used in this study. The non-ideal value is thought to be derived from a combination of polarization errors and a degree of spin-scattering as the excited electrons relax into the quantum dot. The longest storage time recorded here is restricted by the repetition rate of the memory,
Figure 4. Photoluminescence from the positively charged exciton state delayed by $\sim 1 \mu s$ in a single quantum dot with a 2 T perpendicular magnetic field. The three panels show measurements for three differing pump polarizations: (a) horizontal linear (H), (b) right-hand circular (R) and (c) left-hand circular (L). The photoluminescence was measured in the cross-polarized bases as labelled, V indicates vertical linear polarization. The black dotted line is shown as a guide to the eye.

which in turn is limited by the intensity of the emission measured. We expect spin memory to persist for much longer than we can currently measure. Results obtained by using a large ensemble of quantum dots have shown that electron spin information can be maintained for times in excess of 1 ms [8].

The measured asymmetric lineshape of the exciton emission shown in figure 4 is the result of a variable Stark-shift introduced by the applied ac pulse. The turn-on time for the pulse is of the same order as the radiative lifetime of the state ($\sim 1$ ns); hence the energy shift induced by the applied electric field varies throughout the readout cycle. The turn-on time of the ac pulse is limited by the equipment used in this study. A difference in intensity of a factor of 2 between the laser triggered output and the electronically triggered readout from the $X^+$ state is observed. This is also predominantly caused by the slow turn-on of the ac pulse as it enables some emission from the neutral exciton state, which is not collected in this experiment.

Injecting a pair of holes into the quantum dot as opposed to a single hole prevents the formation of long-lived dark exciton states [19] which could severely limit the readout speed of the device. In this experiment the readout time is limited only by the radiative lifetime of the positively charged exciton. Purcell enhancement [20] of the radiative lifetime of exciton states could provide a means to readout a device operating as demonstrated here on a $<100$ ps timescale. Significant enhancement of the exciton’s radiative lifetime has already been demonstrated in systems providing good exciton-cavity mode coupling [21, 22].

The multimode non-resonant excitation used in the write stage here limits the polarization information transferred from the input pulse to the single electron’s spin. Larger quantum dots, providing more confinement than those used in this study, can be quasi-resonantly pumped through a fast relaxing excited state. This should significantly improve the degree of
exciton-spin polarization transferred to the quantum dot. Improvements in the design of the optical cavity should also allow near-deterministic single photon absorption and emission [23]. Single quantum dots could therefore prove to be a useful interface between single photons and electrons finding application in quantum information processing. The difference in energy between the input and output photons in the operation of this device is a very useful feature, it allows the two photons to be spectrally separated and the device to operate with a significant input bandwidth. This could allow the transfer of quantum information between systems operating at different wavelengths.

Acknowledgments

This work was partially funded by the EU projects QAP and SANDiE, and by the EPSRC through the IRC for Quantum Information Processing.

References

[1] Bennett C H and Brassard G 1984 Proc. IEEE Int. Conf. on Computers, Systems and Signal Processing pp 175–9
[2] Cleve R and Buhrman H 1997 Phys. Rev. A 56 1201–4
[3] Knill E, Laflamme R and Milburn G J 2001 Nature 409 46–52
[4] Monroe C, Meekhof D M, King B E, Itano W M and Wineland D J 1995 Phys. Rev. Lett. 75 4714–7
[5] Bimberg D, Grundmann M and Ledentsov N N 1999 Quantum Dot Heterostructures (Chichester: Wiley)
[6] Michler P, Kiraz A, Becher C, Schoenfeld W V, Petroff P M, Zhang L, Hu E and Imamoglu A 2000 Science 290 2282–5
[7] Yuan Z, Kardynal B E, Stevenson R M, Shields A J, Lobo C J, Cooper K, Beattie N S, Ritchie D A and Pepper M 2001 Science 295 102–5
[8] Kroutvar M, Ducommun Y, Heiss D, Bichler M, Schuh D, Abstreiter G and Finley J J 2004 Nature 432 81–4
[9] Reithmaier J P, Sek G, Löffler A, Hofmann C, Kuhn S, Reitzenstein S, Keldysh L V, Kulakovskii V D, Reinecke T L and Forchel A 2004 Nature 432 197–200
[10] Yoshie T, Scherer A, Hendrickson J, Khitrova G, Gibbs H M, Rupper G, Ell C, Shchekin O B and Deppeet D G 2004 Nature 432 200–3
[11] Peter E, Senellart P, Martrou D, Lemaitre A, Hours J, Gerard J M and Bloch J 2005 Phys. Rev. Lett. 95 067401
[12] Blakesley J C, See P, Shields A J, Kardynal B E, Atkinson P, Farrer I and Ritchie D A 2005 Phys. Rev. Lett. 94 067401
[13] Stevenson R M, Young R J, Atkinson P, Cooper K, Ritchie D A and Shields A J 2006 Nature 439 179–82
[14] Young R J, Stevenson R M, Atkinson P, Cooper K, Ritchie D A and Shields A J 2006 New J. Phys. 8 29
[15] Akopian N, Lindner N H, Poem E, Berlatzky Y, Avron J and Gershoni D 2006 Phys. Rev. Lett. 96 130501
[16] Benisty H, Neve H D and Weisbuch C 1998 IEEE J. Quantum Electron. 34 1612
[17] Bayer M, Ortna G, Stern O, Kuther A, Gorbunov A A, Forchel A, Hawrylak P, Fafard S, Hinzer K, Reinecke T L, Walck S N, Reithmaier J P, Klopf F and Schäfer F 2002 Phys. Rev. B 65 195315
[18] Landin L, Miller M S, Pisto-M E, Pryor C E and Samuelson L 1998 Science 280 262–4
[19] Besombes L, Kheng K and Martrou D 2000 Phys. Rev. Lett. 85 425–8
[20] Purcell E M 1946 Phys. Rev. 72 59
[21] Englund D, Fattal D, Waks E, Solomon G, Zhang B, Nakaoka T, Arakawa Y, Yamamoto Y and Vučković J 2005 Phys. Rev. Lett. 95 013904
[22] Gevaux D G, Bennett A J, Stevenson R M, Shields A J, Atkinson P, Griffiths J, Anderson D, Jones G A C and Ritchie D A 2006 Appl. Phys. Lett. 88 131101
[23] Press D, Götzing S, Reitzenstein S, Hours J, Löffler A, Kamp M, Forchel A and Yamamoto Y 2007 Phys. Rev. Lett. 98 117402

New Journal of Physics 9 (2007) 365 (http://www.njp.org/)