Energy-Tunable Quantum Dot with Minimal Fine Structure Created by Using Simultaneous Electric and Magnetic Fields

M. A. Pooley
Toshiba Research Europe Limited, Cambridge Research Laboratory, 208 Science Park, Milton Road, Cambridge, CB4 0GZ, U.K. and Cavendish Laboratory, J. J. Thomson Avenue, Cambridge, CB3 0HE, U.K.

A. J. Bennett, R. M. Stevenson, and A. J. Shields
Toshiba Research Europe Limited, Cambridge Research Laboratory, 208 Science Park, Milton Road, Cambridge, CB4 0GZ, U.K.

I. Farrer and D. A. Ritchie
Cavendish Laboratory, Cambridge University, J. J. Thomson Avenue, Cambridge, CB3 0HE, U.K.

The neutral biexciton cascade of single quantum dots is a promising source of entangled photon pairs. The character of the entangled state is determined by the energy difference between the excitonic eigenstates, known as fine structure splitting (FSS). Here we reduce the magnitude of the FSS by simultaneously using two independent tuning mechanisms, in-plane magnetic field and vertical electric field. We observe that there exists a minimum possible FSS in each QD which is independent of these tuning mechanisms. However, with simultaneous application of electric and magnetic fields we show the FSS can be reduced to its minimum value as the energy of emission is tuned over several meV with a 5T magnetic field.

Semiconductor quantum dots (QDs) confine carriers to nano-sized regions, resulting in the creation of a discrete set of energy levels. Optical transitions between these states in InGaAs QDs have many applications in the fields of quantum optics and optical quantum computation. In particular, the radiative decay of the biexciton state ($|X_2\rangle$) via the exciton state ($|X_1\rangle$), has generated significant interest as a source of on-demand entangled photon pairs\cite{1,2}. This decay process, $|X_2\rangle \rightarrow |X_1\rangle \rightarrow |0\rangle$, is split into two separate paths by the fine-structure splitting (FSS)\cite{3}. A finite value of $|s|$ results in the evolution of the $|X_1\rangle$ state during the time between the two emission events, leading to a time-dependent phase between the two components of the emitted two-photon state\cite{4}. Therefore, for applications which require a known input state, such as photonic quantum computing operations, it is desirable to reduce or eliminate $|s|$.

This has motivated research into methods to manipulate the FSS, including the application of piezoelectric strain\cite{5,6}, intense coherent lasers\cite{7,8}, magnetic fields\cite{9,10}, and electric fields\cite{11,12}. Alternatively, several groups are pursuing the growth of dots under particular conditions that naturally give rise to minimal fine-structure: by targeting particular emission energies on the (100) surface of GaAs \cite{2} or the (111)A surface of GaAs \cite{17}. However, whilst such methods are effective at reducing $|s|$, several studies report coherent coupling between the two exciton eigenstates which results in a minimum value, $s_0$ \cite{6,14,15,18,20}.

The use of an electric field orientated parallel to the sample growth direction is particularly promising, as it allows $|s|$ to be varied over a range on the order of 100 $\mu$eV and permits individual QDs to be independently addressed using multiple electric contacts. In certain samples it is possible to minimise $|s|$ in a significant proportion of QDs from an ensemble. However, the value of $s_0$ varies between QDs and is often non-zero.

Recent theoretical and experimental studies have explored the possibility of manipulating $s_0$ via the simultaneous application of two independent tuning mechanisms. It has been proposed that $s_0$ can be reduced to $\sim 0.1$ $\mu$eV in all InGaAs/GaAs QDs using two combined strain fields\cite{20}. Also, it has been demonstrated that the FSS can be eliminated in such QDs with simultaneous application of a strain field and an electric field\cite{10}. However, in both cases a single minimum $s_0$ is obtained at a particular combination of the two tuning parameters, restricting the emission to a single energy when $s_0 = 0$. In this letter we present a method which allows the emission energy to be varied whilst maintaining the FSS at its minimum value. This is achieved via the use of a Voigt magnetic field, orientated in the plane of the sample, in conjunction with an electric field which is applied parallel to the sample growth direction. We show that, by pre-selection of a QD with sufficiently small $s_0$, this technique may be suitable for the creation of an on-demand ‘energy-tunable’ source of entangled photons. Such a development is pre-requisite for building networks of multiple QDs, connected by photonic interference.

The devices used for this work are p-i-n diodes, as detailed elsewhere\cite{21}, in which the QDs are placed at the center of the intrinsic region, between two AlGaAs/GaAs superlattice tunnel barriers, inside a planar cavity with 2 and 13 periods above and below the QDs, respectively.

\*anthony.bennett@crl.toshiba.co.uk
These devices allow the application of an electric field in the sample growth direction, leading to large Stark shifts in the transition energies (Figure 1(a)). The transition linewidths remain below the resolution of our spectrometer across this electric field range \(21\). We choose to apply magnetic field perpendicular to the growth direction, parallel to the [110] crystal axis (Voigt configuration) using a superconducting magnet, as this has previously been shown to reduce the FSS in certain dots \(10\). In contrast, a magnetic field parallel to the growth direction always leads to an increase in the FSS \(8\). Figure 1(b) shows a schematic diagram of the device structure along with the orientation of the electric and magnetic fields. The FSS is extracted from the energy difference between exciton and biexciton transitions recorded as a function of polarization angle (as described in \(14\)). This technique enables the fine structure to be measured with an error of \(\pm 0.5 \mu eV\), using a spectrometer with a resolution of \(\sim 25 \mu eV\). However, at small values of the FSS it is not possible to separate the polarisation properties of the two neutral eigenstates but the total emission is isotropic.

The behavior of \(|s|\) as a function of electric field, \(F\), is described by a hyperbola given by

\[
|s| = \sqrt{\gamma^2 (F - F_0)^2 + s_0^2},
\]

where \(\gamma\) is the rate at which \(|s|\) varies with \(F\) in the absence of coupling effects, and \(F_0\) is the electric field required to minimise \(|s|\) \(14\). Figure 2(a) shows \(|s|\) as a function of \(F\) for five different magnitudes of Voigt magnetic field, \(B\), for an example QD with an \(s_0\) of \(2.0 \pm 0.2 \mu eV\). The solid lines in Figure 2(a) show least-squares fits to the data using equation 1 from which the data points and errors for \(F_0\), \(s_0\) and \(\gamma\) are extracted (Figure 2(b)-(d)). These figures show that for a given magnetic field it is possible to minimize the FSS at a certain electric field \(F_0\), but that \(\gamma\) and \(s_0\) are unchanged by \(B\). The constant value of \(\gamma\) suggests that a Voigt magnetic field does not affect the difference in permanent \(z\) dipole moment between the two exciton eigenstates \(22\).

This variation of \(F_0\) as a function of \(B\) can be explained by considering the additional contribution to the fine structure splitting due to the Voigt magnetic field, along with the observation that the coupling strength \(s_0\) between the eigenstates is independent of this field. The magnetic field induces an additional splitting, \(\Delta s\), between the two exciton eigenstates which is well approximated by \(\Delta s = \kappa B^2\), where \(\kappa\) is dependent on the in-plane anisotropy of the QD along with the g-factors of the confined carriers \(10, 23, 24\). Thus, in the presence of a Voigt magnetic field, the magnitude of \(F\) required to minimize \(|s|\) is increased. The value of \(F_0\) is then given by

\[
F_0 (B) = F_0 (0) - \frac{\kappa}{\gamma} B^2,
\]

where \(F_0(0)\) is the value of \(F_0\) in the absence of the magnetic field and the second term in this equation is the change in electric field required to remove the additional, magnetically induced, component of the fine structure splitting. Figure 2(b) shows \(F_0(B)\) as a function of magnetic field fit with equation 2 from which a value of \(\kappa = 0.45 \pm 0.02 \mu eV T^{-2}\) is extracted.

The effect of simultaneous application of both the electric and magnetic field is found by substituting equation 2 into equation 1. Figures 2(e)-(g) show \(|s|\) as a function of \(B\) for three different values of \(F\). There is good agreement between the experimental measurements and the model. The value of \(|s|\) is either increased or decreased by the application of \(B\), depending on the relative sign of \(\kappa\) and that of the FSS at zero magnetic field, \(s(B = 0)\). For the QDs studied here \(\kappa > 0\), in figure 2(e) \(s(B = 0) > 0\) leading to an increase in \(|s|\) with \(B\); whereas in figure 2(f) \(s(B = 0) < 0\), resulting in a reduction of \(|s|\). This behavior is similar to that reported in \(10\). However, Figure 2(f) shows data at -301 kV/cm where \(B\) causes the fine structure to pass through a minimum value, \(s_0\). This behavior has not been observed before, and indicates that the avoided crossing in \(s\) is a fundamental property of these QDs and not an artifact of the tuning mechanism \(13, 25\).

Figure 3 shows data from the neutral cascade for a second QD with \(s_0 = 0.6 \pm 0.5 \mu eV\), which is below the FSS of those QDs previously shown to generate entangled photon pairs \(2, 14\). The behavior is similar to that of all dots we have studied, such as the example in Figure 2 with a larger \(s_0\). Polarized spectra from this dot are shown in...
At higher magnetic fields the mixing of dark and bright exciton states [23] results in the appearance of weak emissions and widths are observed over this range of magnetic fields. The value of $|s|$ as a function of $F$ for six values of $B$ was measured and used to determine $s_0$ (Figure 3) along with the emission energy of the two bright neutral transitions at $F_0(B)$, (figure 3(b)). The energy of the photons emitted from the bright neutral transitions depends on $F$, due to the electric field dependent Stark shift described by

$$E = E_0 - pF + \beta F^2,$$

where $E_0$ is the energy in the absence of an electric field, $p$ is the component of the dipole moment which is parallel with $F$, and $\beta$ is the polarisability. Therefore, the ability to tune the value of $F_0$ allows the energy of the photons in the emitted two-photon state to be varied whilst maintaining the FSS at a minimum value of $|s| = s_0$. The energy of the photons emitted at $F = F_0$ is found by combining equations 2 and 3.

From the parabolic shift of the emission energy, Stark shift parameters of $p = -6.1 \pm 0.4 \mu eV \text{ cm}^{-1}$ and $\beta = 0.15 \pm 0.3 \mu eV \text{ cm}^{-2}\text{ kV}^{-2}$ are obtained.

The range over which the photon energy can be tuned is dependent on two factors: the maximum electric field which can be applied without quenching the optical activity of the QD; and the maximum available magnetic field which can be applied. For the work presented here, the tuning range was restricted by the latter. However, the tuning range is quadratically increased by considering magnetic fields greater than those available in this
study. For the devices studied in this work, the maximum magnitude of electric field which can be applied whilst preserving optical emission is \( F_{\text{max}} \sim -430 \text{ kV cm}^{-1} \). At \( B = 5T \) the electric field required to minimise \( |s| \) is \( F_0(B = 5T) = -151 \text{ kV cm}^{-1} \), which is well below this value. It is possible to increase the tuning range by using a larger magnetic field until \( F_0(B) \) reaches \( F_{\text{max}} \) which occurs at \( B = 11 T \). Figure 9(b) shows the emission energy of the two neutral transitions at \( F = F_0 \) for \( B \) up to 12 T. The maximum tuning range possible with these devices, which is achieved with a magnetic field of \( B = 11 T \), is 22.5 meV and 25.4 meV for the \( |X_2 \rangle \rightarrow |X_1 \rangle \) and \( |X_1 \rangle \rightarrow |0 \rangle \) transitions respectively. This range could be further improved by increasing the amount of AlGaAs in the device barrier layers, thus reducing carrier tunneling further and increasing \( F_{\text{max}} \).

In conclusion, we have presented a method of manipulating \( |s| \) using simultaneous application of a Voigt geometry magnetic field and an orthogonal electric field. In particular, we have demonstrated that this method may be suitable for the generation of energy-tunable entangled photon pairs, using QDs selected to have small \( s_0 \). Experimentally we achieve a tuning range of a few meV with a 5T magnetic field but show this could be extended to tens of meV with an 12T magnet. We expect our technique to find application to dots emitting at energies where more efficient detectors are available and to dots emitting at wavelengths compatible with the absorption minima in optical fibres. A promising future development of this method would be to incorporate an applied strain field as a third independent tuning mechanism. As simultaneous application of strain and electric field has been demonstrated to reduce \( s_0 \) to zero in any QD, it removes the requirement to pre-select QDs with small \( s_0 \).

[1] O. Benson, C. Santori, M. Pelton, and Y. Yamamoto, Phys. Rev. Lett. 84, 2513 (2000)
[2] C. L. Salter, R. M. Stevenson, I. Farrer, C. A. Nicoll, D. A. Ritchie, and A. J. Shields, Nature 465, 594 (2010)
[3] M. Bayer, G. Ortner, O. Stern, A. Kuther, A. A. Gorbunov, A. Forchel, P. Hawrylak, S. Fafard, K. Hinzer, T. L. Reinecke, S. N. Walck, J. P. Reithmaier, F. Klopf, and F. Schafer, Phys. Rev. B 65, 195315 (2002)
[4] R. M. Stevenson, A. J. Hudson, A. J. Bennett, R. J. Young, C. A. Nicoll, D. A. Ritchie, and A. J. Shields, Phys. Rev. Lett. 101, 170501 (2008)
[5] S. Seidl, M. Kroner, A. Hogele, K. Karrai, R. J. Warburton, A. Badolato, and P. M. Petroff, Appl. Phys. Lett. 88, 203113 (2006)
[6] G. Bryant, M. Zielinski, J. J. W. Malkova, and Sims, and J. Aizpurua, Phys. Rev. Lett. 105, 067404 (2010)
[7] G. Sundt, L. Robledo, A. Hogele, S. Falt, and A. Imamoglu, Phys. Rev. Lett. 100, 177401 (2008)
[8] A. Muller, W. Fang, J. Lawall, and G. S. Solomon, Phys. Rev. Lett. 103, 217402 (2009)
[9] M. Bayer, A. Kuther, A. Forchel, A. Gorbunov, V. B. Timofeev, F. Schaper, J. P. Reithmaier, T. L. Reinecke, and S. N. Walck, Phys. Rev. Lett. 82, 1748 (1999)
[10] R. M. Stevenson, R. J. Young, P. See, D. G. Gevaux, K. Cooper, P. Atkinson, I. Farrer, D. A. Ritchie, and A. J. Shields, Phys. Rev. B 73, 033306 (2006)
[11] M. M. Vogel, S. M. Ulrich, R. Hafenbrak, P. Michler, L. Wang, A. Rastelli, and O. G. Schmidt, Appl. Phys. Lett. 91, 051904 (2007)
[12] B. D. Gerardot, S. Seidl, P. A. Dalgarno, R. J. Warburton, D. Granados, J. M. Garcia, K. Kowalik, O. Krebs, K. Karrai, A. Badolato, and P. M. Petroff, Appl. Phys. Lett. 90, 041101 (2007)
[13] K. Kowalik, O. Krebs, A. Lemaître, B. Eble, A. Kudelski, P. Voisin, S. Seidl, and J. A. Gaj, Appl. Phys. Lett. 91, 183104 (2007)
[14] A. J. Bennett, M. A. Pooley, R. M. Stevenson, M. B. Ward, R. B. Patel, A. B. de la Giroday, N. Skold, I. Farrer, C. A. Nicoll, D. A. Ritchie, and A. J. Shields, Nat. Phys. 6, 947 (2010)
[15] K. Kowalik, O. Krebs, P. Senellart, A. Lemaître, B. Eble, A. Kudelski, J. Gaj, and P. Voisin, Physica Status Solidi (c) 3, 3890 (2006)
[16] S. Marcet, K. Ohtani, and H. Ohno, Appl. Phys. Lett. 96, 101117 (2010)
[17] T. Kuroda, T. Mano, N. Ha, H. Nakajima, H. Kuman, B. Urbaszek, M. Jo, M. Abbarchi, Y. Sakuma, Y. Sakoda, I. Suemune, X. Marie, and T. Amand, Physical Review B 88, 041306 (R) (2013).
[18] R. Singh and G. Bester, Phys. Rev. Lett. 104, 196803 (2010)
[19] R. Trotta, E. Zallo, C. Ortiz, P. Atkinson, J. D. Plumphof, J. van den Brink, A. Rastelli, and O. G. Schmidt, Phys. Rev. Lett.109, 147401 (2012)
[20] J. Wang, M. Gong, G.-C. Guo, and L. He, Appl. Phys. Lett. 101, 063114 (2012)
[21] A. J. Bennett, R. B. Patel, J. Skiba-Szymanska, C. A. Nicoll, I. Farrer, D. A. Ritchie, and A. J. Shields, Appl. Phys. Lett. 97, 031104 (2010)
[22] J. J. Finley, M. Sábatil, P. Vogl, G. Abstreiter, R. Oulton, A. I. Tartakovskii, D. J. Mowbray, M. S. Skolnick, S. L. Liew, A. G. Cullis, and M. Hopkinson, Phys. Rev. B 70, 201308 (2004)
[23] M. Bayer, O. Stern, A. Kuther, and A. Forchel, Phys. Rev. B 61, 7273 (2000)
[24] A. J. Bennett, M. A. Pooley, Y. Cao, N. Skild, I. Farrer, D. A. Ritchie, and A. J. Shields, Nature Communications 4, 1522 (2013).
[25] J. D. Plumphof, V. Krápek, F. Ding, K. D. Jøns, R. Hafenbrak, P. Klenovsky, A. Herkelz, K. Dörr, P. Michler, A. Rastelli, and O. G. Schmidt, Phys. Rev. B 83, 121302 (2011)
[26] M. B. Ward, M. Dean., M. Stevenson, A. J. Bennett, D. Ellis, K. Cooper, I. Farrer, C. Nicoll, D. Ritchie, and A. J. Shields, Nature Communications 5, 3316 (2014).