Observation of anticrossings in the exciton state of single quantum dots via electrical tuning of the fine-structure splitting

M A Pooley, A J Bennett, R M Stevenson, M B Ward, R B Patel, A Boyer de la Giroday, N Sköld, I Farrer, C A Nicoll, D A Ritchie and A J Shields

1 Toshiba Research Europe Limited, Cambridge Research Laboratory, 208 Science Park, Milton Road, Cambridge, CB4 0GZ, UK
2 Cavendish Laboratory, Cambridge University, J J Thomson Avenue, Cambridge, CB3 0HE, UK

E-mail: map57@cam.ac.uk

Abstract. Single quantum dots have many potential applications across the field of quantum computation, ranging from the generation of single photons or entangled photon pairs to the storage and manipulation of qubits. Single InAs quantum dots are optically active and thus can be used as an interface between photonic flying qubits and spin-based stationary qubits. Incorporating single InAs quantum dots into semiconductor devices allows the stationary qubits to be manipulated, making this system a promising candidate for quantum computation. It is well known that the exciton state of quantum dots is split into two polarisation dependent energy eigenstates, separated by a fine-structure splitting (\(s\)) [2]. This system has been the subject of much research, particularly in the field of quantum computation, however until recently the two eigenstates have been considered independent [3–7]. We have demonstrated that it is possible to electrically induce coherent coupling of the exciton states using a vertically applied electric field [1]. The electric field tunes \(s\) by Stark-shifting the two eigenstates at different rates. When the two eigenstates are brought close together an anticrossing is observed indicating coherent coupling. The magnitude of \(s\) is important because it is easier to observe entangled photon generation from the biexciton-exciton-empty cascade when \(s\) is small. It is therefore often desirable to work with QDs that have as small \(s\) as possible, however it is not always possible to grow QDs that naturally have small \(s\). This has motivated many other techniques for tuning \(s\), including the use of magnetic field [2, 3], lateral

1. Introduction

It is well known that the neutral exciton states of quantum dots (QDs) are split into two polarisation dependent energy eigenstates, separated by a fine-structure splitting (\(s\)) [2]. This system has been the subject of much research, particularly in the field of quantum computation, however until recently the two eigenstates have been considered independent [3–7]. We have demonstrated that it is possible to electrically induce coherent coupling of the exciton states using a vertically applied electric field [1]. The electric field tunes \(s\) by Stark-shifting the two eigenstates at different rates. When the two eigenstates are brought close together an anticrossing is observed indicating coherent coupling. The magnitude of \(s\) is important because it is easier to observe entangled photon generation from the biexciton-exciton-empty cascade when \(s\) is small. It is therefore often desirable to work with QDs that have as small \(s\) as possible, however it is not always possible to grow QDs that naturally have small \(s\). This has motivated many other techniques for tuning \(s\), including the use of magnetic field [2, 3], lateral
electric field [6–8], and strong coherent lasers [4, 5]. However, those that have been able to minimize \( s \) have not seen an anticrossing. The application of vertical electric field is arguably the most practical technique for tuning \( s \), as it can be achieved by simply placing the QDs in the center of a p-i-n diode and allows easy dynamic control of the applied field using standard voltage sources.

The magnitude of the anticrossing \( (s_0) \) is the minimum value to which \( s \) can be reduced using this technique, and so determines how suitable a particular QD is for use as an entangled photon pair source. We have measured an ensemble of QDs and found that \( s_0 \) varies from dot to dot, with some dots having sufficiently small \( s_0 \) to allow entangled photon pairs to be observed.

### 2. Sample design and methods

Application of a vertical electric field can lead to quenching of the QD emission by increasing the rate at which carriers tunnel out of the QD; once the tunneling time is comparable with the time for radiative recombination the emission is drastically reduced. To overcome this problem InAs QDs are placed inside a 10nm thick two-dimensional GaAs quantum well which is sandwiched between two AlAs/GaAs short-period superlattices equivalent to Al\(_{0.75}\)Ga\(_{0.25}\)As. This increases the carrier confinement in the region of the QDs and limits the rate at which carriers can tunnel away. Distributed bragg reflectors are used to create a weak planar microcavity, with 14/4 periods on the bottom/top of the device. The structure is doped to create a p-i-n device with a \( d = 140\)nm thick intrinsic region centered around the dot layer. The electric field applied to the QDs can be calculated as \((V - V_{bt})/d\), where \( V_{bt} \) is the built-in potential which for these samples is 2.2V. An opaque aluminum film is deposited on the surface and patterned with arrays of \( \mu \)m diameter apertures through which optical excitation and emission collection occurs. These apertures allow individual QDs to be optically addressed. This sample design enables very large electric fields to be applied to the QDs before emission is quenched and so allows the observation of giant Stark effects [9]. Figures 1 and 2 show the device structure and a typical plot of the photoluminescence from a single QD as a function of applied electric field.

![Figure 1](image1.png)  
**Figure 1.** Diagram of device structure. The quantum dots are embedded in a 2D quantum well inside a weak planar microcavity.

![Figure 2](image2.png)  
**Figure 2.** Example of photoluminescence as a function of electric field for a single quantum dot.

The QDs were optically excited with a continuous-wave 850nm diode laser, which creates carriers both in the QDs and the wetting layer. The spectra from single QDs is well understood
Figure 3. Example of photoluminescence as a function of polarisation angle. There are two clear energy eigenstates separated by 90°.

and the spectral lines can be identified as the biexciton ($X_2$), exciton ($X$), and two charged excitons ($X^+$ and $X^-$), where this notation refers to the initial state of each transition.

Measurements of $|s|$, and the eigenstate orientation ($\theta$) were obtained by recording polarised spectra for a range of polarisation angles between 0° and 180°. The value of $|s|$ is given by the energy separation of the two orthogonal eigenstates. Figure 3 shows spectra taken from a QD with large $|s|$ where the two different eigenstates can clearly be resolved; light with $\theta = 0^\circ$ has different energy from that with $\theta = 90^\circ$. The linewidth of the spectral peaks is dominated by the resolution of the spectrometer. In cases where $|s|$ cannot be clearly resolved a fitting procedure is used to extract the energy of peak emission at each angle; the peak energies are fit with a sinusoidal function the amplitude of which yields $|s|$. The angles at which the emission from each state is maximum are used to extract their orientation and thus obtain $\theta$. Small energy shifts caused by rotation of the polarisation optics are eliminated by finding the difference between the biexciton and exciton transition energies giving submicroelectronvolt accuracy [3, 10].

3. Results

As can be seen in Figure 2, the application of a vertical electric field ($F$) induces a Stark shift of the emission lines, this shift takes the usual form,

\[ E = E_0 - pF + \beta F^2 \]  

where $p$ is the $z$-component of the permanent dipole moment and $\beta$ is the polarizability. Far from the anticrossing the eigenstates tend to align with the [110] and [1\bar{1}0] crystalline axis and can be labeled H (V) for horizontal (vertical) polarization. Away from their anticrossings $s$ shifts linearly with electric field at a rate of $\gamma = -0.285 \pm 0.019\mu$eV kV$^{-1}$ cm (see figure 4). We attribute this to a slight difference in the confinement energy of the two eigenstates, which results in each eigenstate having a different $p$; thus $\gamma$ is given by $\gamma = p_H - p_V$.

As $|s|$ is tuned to a minimum an anticrossing of the two eigenstates is observed, which is a clear indication of coherent coupling. This behavior is well described by a coupling matrix of the same form as that used to describe strong light-matter coupling [11] and anticrossings in the states of molecular systems [12],

\[ E \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} = \begin{pmatrix} E_0 & s_0/2 \\ s_0/2 & E_0 - \gamma(F - F_0) \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} \]  

where $F_0$ is the field at which the minimal splitting occurs.
Far from the anticrossing, when $\gamma(F - F_0) >> s_0$, the states are aligned to the crystalline axis with H and V polarized eigenstates. Close to $(F - F_0) = 0$ the eigenstates are a coherent mixture with components $\sin \theta$ and $\cos \theta$ of H and V polarized light. The orientation angle $\theta$ is thus the actual angle of the eigenstates relative to the crystalline axis. The eigenvalues, $E_{\pm}$, and eigenvectors of equation 2 are

$$E_{\pm} = E_0 - \frac{\gamma(F - F_0)}{2} \pm \frac{1}{2} \sqrt{\gamma^2(F - F_0)^2 + s_0^2}$$

$$\theta_{\pm} = \pm \tan^{-1} \left( \frac{s_0}{\gamma(F - F_0) \pm (E_- - E_+)} \right)$$

Although there are two degenerate solutions, $\theta_{\pm}$, which describe either clockwise or anticlockwise rotation of the eigenstates, we observe that each QD only ever rotates in one direction. Our measurements are consistent with the ensemble of QDs having no preferred rotation direction.

Figure 5 shows data from three QDs, the data is in excellent agreement with the simple model.

We now discuss how this system could find applications as a solid state quantum memory. Clearly for a memory coherent writing of photon polarization to the spin-state of the exciton is required, and we show this is possible with high fidelity. In addition, we show that the state remains stored coherently on a timescale of several times the radiative lifetime. In order to find practical use this storage time must be increased substantially.

Quasi-resonant excitation of a QD with a linearly polarized laser can be used to write the polarization state of the photons onto the spin state of the carriers created in the QD. If the QD is excited into a superposition of the two polarization eigenstates, time-resolved measurements of the emission from a superposition state yield coherent oscillations. The frequency of the oscillations is given by $|s|/\hbar$, and the visibility is maximal when exciting and measuring maximum superposition states. Figure 6 shows such oscillations for a QD tuned to three different values of $|s|$. The oscillations are visible for longer than the radiative lifetime allowing storage of the qubit for $\sim 3$ns. In addition, it is possible to dynamically vary $s$ whilst...
Figure 5. Measurements of fine structure splitting ($|s|$) and eigenstate orientation ($\theta$) as a function of electric field for three quantum dots. The red lines show fits to the data using equations 3 and 4.

Figure 6. Time-resolved measurements of the exciton emission from a dot tuned to three different values of fine structure splitting showing coherent oscillations of the spin-state. The dot was excited into a maximum superposition state and measurement was along the same orientation as excitation. Data points are shown as solid black circles, the red lines are a guide to the eye.

the qubit is stored in the QD. This allows the orientation of the eigenstates to be varied and the coherent oscillations to be controlled [13].

4. Conclusions
We have used vertical electric fields to tune the fine-structure splitting of single QDs over a very large range of more than 100$\mu$eV. Tuning $s$ close to zero has resulted in the observation of an anticrossing of the two polarisation eigenstates, which is attributed to coherent coupling of the
states. This has important implications as the magnitude of the anticrossing determines if a particular QD is suitable as a practical entangled photon pair source.

This system also has potential applications in quantum memory and quantum computation. The optically active QDs allow flying photonic qubits to be easily transferred to and from the solid state, where manipulations or logical operations can be performed.

References

[1] Bennett A J, Pooley M A, Stevenson R M, Ward M B, Patel R B, de la Giroday A B, Skold N, Farrer I, Nicoll C A, Ritchie D A and Shields A J 2010 Nat Phys advance online publication – ISSN 1745-2481 URL http://dx.doi.org/10.1038/nphys1780

[2] Bayer M, Kuther A, Forchel A, Gorbunov A, Timofeev V B, Schafer F, Reithmaier J P, Reinecke T L and Walck S N 1999 Phys. Rev. Lett. 82 1748– URL http://link.aps.org/doi/10.1103/PhysRevLett.82.1748

[3] Stevenson R M, Young R J, See P, Gevaux D G, Cooper K, Atkinson P, Farrer I, Ritchie D A and Shields A J 2006 Phys. Rev. B 73 033306– URL http://link.aps.org/doi/10.1103/PhysRevB.73.033306

[4] Jundt G, Robledo L, Hogele A, Falt S and Imamoglu 2008 Phys. Rev. Lett. 100 177401– URL http://link.aps.org/doi/10.1103/PhysRevLett.100.177401

[5] Muller A, Fang W, Lawall J and Solomon G S 2009 Phys. Rev. Lett. 103 217402– URL http://link.aps.org/doi/10.1103/PhysRevLett.103.217402

[6] Kowalik K, Krebs O, Lemaitre A, Eble B, Kudelski A, Voisin P, Seidl S and Gaj J A 2007 Appl. Phys. Lett. 91 183104–3 URL http://link.aip.org/link/?APL/91/183104/1

[7] Gerardot B D, Seidl S, Dalgarno P A, Warburton R J, Granados D, Garcia J M, Kowalik K, Krebs O, Karrai K, Badolato A and Petroff P M 2007 Appl. Phys. Lett. 90 041101–3 URL http://link.aip.org/link/?APL/90/041101/1

[8] Vogel M M, Ulrich S M, Hafenbrak R, Michler P, Wang L, Rastelli A and Schmidt O G 2007 Applied Physics Letters 91 051904 URL http://dx.doi.org/10.1063/1.2761522

[9] Bennett A J, Patel R B, Skiba-Szymanska J, Nicoll C A, Farrer I, Ritchie D A and Shields A J 2010 Appl. Phys. Lett. 97 031104–3 URL http://link.aip.org/link/?APL/97/031104/1

[10] Young R J, Stevenson R M, Shields A J, Atkinson P, Cooper K, Ritchie D A, Groom K M, Tartakovskii A I and Skolnick M S 2005 Phys. Rev. B 72 113305– URL http://link.aps.org/doi/10.1103/PhysRevB.72.113305

[11] Khitrova G, Gibbs H M, Kira M, Koch S W and Scherer A 2006 Nat Phys 2 81–90 ISSN 1745-2473 URL http://dx.doi.org/10.1038/nphys227

[12] Stinaff E A, Scheibner M, Bracker A S, Ponomarev I V, Korenev V L, Ware M E, Doty M F, Reinecke T L and Gammon D 2006 Science 311 636–639 URL http://www.sciencemag.org/cgi/content/abstract/311/5761/636

[13] Boyer de la Giroday A, Bennett A J, Pooley M A, Stevenson R M, Ward M B, Patel R B, Skold N, Farrer I, Nicoll C A, Ritchie D A and Shields A J accepted for publication in Phys Rev B Rapid Communications, arXiv:1011.2641v1