

Quantum-confined Stark effect in a single InGaN quantum dot under a lateral electric field

James W. Robinson, James H. Rice, Kwan H. Lee, Jong H. Na, and Robert A. Taylor^{a)}
Department of Physics, University of Oxford, Parks Road, Oxford, OX1 3PU, United Kingdom

David G. Hasko
Microelectronics Research Centre, University of Cambridge, Madingley Road, Cambridge, CB3 0HE, United Kingdom

Rachel A. Oliver, Menno J. Kappers, and Colin J. Humphreys
Department of Materials, University of Cambridge, Pembroke Street, Cambridge, CB2 3QZ, United Kingdom

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

(Received 29 November 2004; accepted 11 April 2005; published online 16 May 2005)

The effect of an externally applied lateral electric field upon an exciton confined in a single InGaN quantum dot is studied using microphotoluminescence spectroscopy. The quantum-confined Stark effect causes a shift in the exciton energy of more than 5 meV, accompanied by a reduction in the exciton oscillator strength. The shift has both linear and quadratic terms as a function of the applied field. © 2005 American Institute of Physics. [DOI: 10.1063/1.1935044]

Semiconductor quantum dots (QDs) are able to confine carriers in all three spatial dimensions. These zero-dimensional nanostructures therefore display discrete energy levels analogous to those of an atom. A number of physical phenomena observed in atomic physics have also been seen in QDs, including the quantum-confined Stark effect (QCSE).^{1,2} For example, the QCSE has been observed in self-assembled QDs through spectral diffusion,^{3,4} i.e., the occurrence of random spontaneous shifts in the exciton recombination energy due to trapped carriers inducing temporally fluctuating local electric fields in close proximity to excitons localized within QDs. The study of the QCSE is important both for fundamental physics and in device applications. The QCSE has been applied in ultrafast optoelectronics, for example, in electro-optical modulators. Studying the Stark shift of excitons permits a better understanding of inter-QD charge distribution as the permanent dipole moment created by electron-hole wave function separation produces a linear energy shift with electric field, while polarizability produces a quadratic energy shift with field.

The investigation of QD materials at the single-QD level has allowed a better understanding of the basic physics of these materials.^{5,6} A number of studies characterizing the QCSE in single-self-assembled QDs have shown that the application of an external electric field in the lateral direction permits control of the exciton wave function via the QCSE. Studies on GaAs QDs have demonstrated that the electric field produces an energy shift with linear and quadratic components, arising respectively from permanent dipole moment and polarizability contributions.⁷ This is in contrast to CdSe QDs, for which only a quadratic energy shift with an electric field is observed, the permanent dipole moment contribution being seen to be negligible.^{8,9}

Nitride-based III/V QD nanomaterials (e.g., InGaN and GaN) (Refs. 10–12) possess interesting electronic properties

such as built-in electric fields and high exciton binding energies that are not found in the widely studied arsenide-based III/V family of QDs. In this letter, the effect of applying an external lateral electric field to a single InGaN QD is studied. A shift in the exciton recombination energy (ΔE) and reduction in the Photoluminescence (PL) intensity are observed. Evidence for carrier tunneling arising from the application of the electric field (F) is presented. The field-dependent shift as expressed by $\Delta E(F) = \mu F - \alpha F^2$ is found to possess nonzero linear and quadratic contributions

The InGaN QD sample was grown by metalorganic vapor phase epitaxy.¹⁰ About 10 monolayers of InGaN were grown on 2.8- μm -thick GaN templates, dots formed on this layer were then capped with 7 nm of GaN. The QD density was determined from atomic force microscopy measurements to be $\sim 1 \times 10^{10} \text{ cm}^{-2}$, which allowed for single-QD measurements. The In content was estimated to be 20%–25%. A 100-nm-thick Al layer was evaporated onto the sample surface, and an electrode pattern (consisting of a pair of Al electrodes with a separation of 1 μm) was fabricated by electron-beam lithography and reactive ion etching using SiCl_4 .

Since the cap layer is thin, an electric field perpendicular to the dot layer will exist due to the presence of surface and polarization charges.¹³ Calculation of this field has not been attempted due to the complexity of estimating the surface charge density. For a bias of 10 V applied across the gap between the electrodes, a rough estimate of the additional electric field parallel to the QD layer has been made using a device simulation package (ATLAS, Silvaco International). Neither residual doping nor the potential effect of charged defects were considered. The component of this additional field in the direction parallel to the QD layer varied between a maximum of 0.17 MV cm^{-1} and a minimum of 0.065 MV cm^{-1} . The mean field across the entire gap was estimated to be 0.093 MV cm^{-1} .

For micro-PL (μPL) measurements, the sample was mounted in a cold-finger cryostat that could be cooled to

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

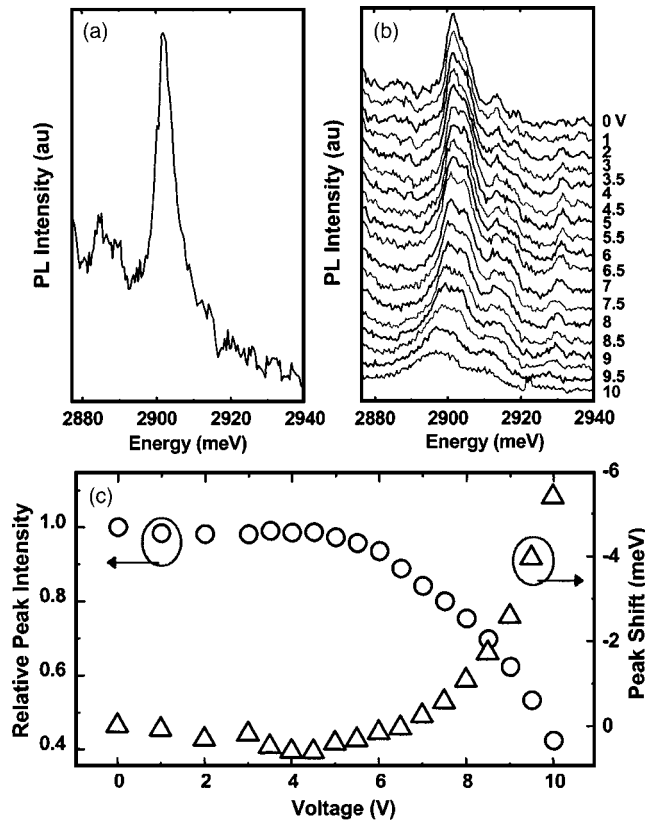


FIG. 1. (a) A μ PL spectrum from a single InGaN QD, recorded at 4.2 K with no externally applied voltage. The integration time was 10 s. The spectrum shows a peak at 2902 meV originating from an optical transition in a single InGaN QD. (b) A series of μ PL spectra recorded sequentially with increasing externally applied voltage, from 0 V to 10 V. The integration time was 10 s in each case. (c) Plots of relative peak intensity and peak shift as a function of applied voltage for the μ PL spectra shown in (b).

4.2 K. A frequency-tripled Ti:Sapphire laser operating at 266 nm was focused through a microscope objective lens to a spot size of $\sim 2 \mu\text{m}$, and the spot was aligned with the electrodes on the sample surface using a charge coupled device (CCD) camera. The μ PL signal was collected by the microscope objective lens, and monitored using a CCD detector, having been dispersed by a 0.3 m monochromator equipped with a 1200 grooves/mm grating.

Figure 1(a) shows a spatially resolved μ PL spectrum from a single InGaN QD, recorded at 4.2 K without an externally applied voltage. The spectrum exhibits a narrow peak originating from exciton recombination. An electric field was then applied in the lateral direction using the electrodes placed on the sample surface. Figure 1(b) shows a series of consecutively recorded μ PL spectra from the QD with increasing field strength. The PL peak redshifts and declines in intensity as the voltage is increased from 0 V to 10 V. The effect of the applied electric field upon the QD is shown in Fig. 1(c), which plots the relative peak intensity and the peak shift as a function of the applied voltage. At 10 V, a peak shift of $\Delta E = 5.4 \text{ meV}$ to lower energy is found. This is in line with previous measurements on epitaxial arsenide-based III/V QDs and selenide-based II/VI QDs, which showed redshifts in exciton recombination energies of up to $\Delta E = 1.1 \text{ meV}$.⁷⁻⁹ This behavior can be readily understood in terms of the QCSE. The electric field forces the electron and hole to opposite sides of the QD and band tilting leads to a reduction in the band gap and thus to a redshift of

the PL. Other mechanisms should also be considered. The application of voltage across a small gap may cause ohmic heating. Sharp exciton recombination peaks in the μ PL spectrum of single InGaN QDs have been observed to broaden, redshift, and merge into the wetting layer background as the temperature rose from 4.2 K to 70 K.¹⁰ The rise in temperature caused shifts of $\Delta E < 3 \text{ meV}$, which is smaller than the shift of $\Delta E = 5.4 \text{ meV}$ observed when the external electric field was applied, indicating that the shifts shown in Fig. 1(b) cannot be explained by local heating effects alone. Spectral diffusion in InGaN QDs (Ref. 4) shows variations in exciton recombination energy due to transient QCSE shifts caused by temporally varying locally generated electric fields (originating from trapped carriers). These shifts in the exciton recombination energy had magnitudes of up to $\Delta E = 1.45 \text{ meV}$ (on a time scale of $\sim 1 \text{ h}$). Such shifts of up to $\Delta E = 1.45 \text{ meV}$ in exciton energy are significantly smaller than the shifts generated by the external lateral electric field [shown in Fig. 1(b)]. Therefore, the contribution of the QCSE shifts associated with spectral diffusion to the shifts in exciton energies seen in this work are not considered to be significant.

The electric-field simulations indicated that close to the electrodes a significant component of the applied electric field was in the direction perpendicular to the sample surface. The QD shown in Fig. 1 may therefore be experiencing a component of an external electric field parallel to its vertical internal electric field. Since the applied lateral field is much stronger than the applied vertical field across the majority of the gap between the electrodes, it is likely that the effect of the lateral component dominates. However, it should be noted that we have not estimated the dot's internal electric field in either the vertical or the lateral direction.

In Fig. 1(c), the intensity of the PL peak drops from a relative value of 1.0 at 0 V to 0.4 at 10 V, in accordance with observations made in other epitaxial QD systems.⁷⁻⁹ The application of the electric field results in a reduction of the oscillator strength because of the decreased electron-hole wave function overlap. An additional process which may also reduce the PL intensity is field-induced carrier tunneling out of the QD. Tunneling may cause nonlinear linewidth broadening in other QD systems.¹⁴ The increase in the linewidth $\Delta\Gamma$ induced by electric field F can be described by:¹⁴

$$\Delta\Gamma = \frac{\pi\hbar^2}{2m^*a^2} \exp\left(-\frac{4\sqrt{2m^*}V_0^{3/2}}{3\hbar eF}\right), \quad (1)$$

where a is the characteristic dimension of the QD; V_0 is the energy offset between the ground state of the electron in the QD and the lateral barrier, and m^* is the reduced electron mass, which may be estimated by assuming an indium content of 20% (Ref. 10) and performing a linear interpolation between the values of m^* for GaN ($0.20m_e$) and for InN ($0.042m_e$),¹⁵ giving $0.17m_e$. Tunneling of holes is neglected because the rate will be much less than for electrons due to their larger effective mass.

Figure 2(a) shows a plot of linewidth against voltage for the QD shown in Fig. 1(b). This plot was fitted with Eq. (1), giving a QD diameter of 2.1 nm. The QDs used in this work have been measured by atomic force microscopy to have an average height of 0.93 nm.¹⁰ The value 2.1 nm obtained is thus a reasonable fit to the measured average QD dimensions. The energy offset V_0 between the ground state of the electron and the lateral barrier was found from the fit to be

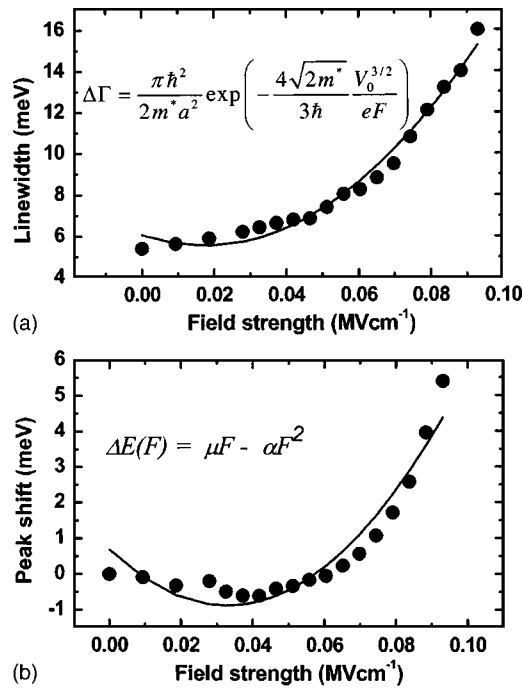


FIG. 2. Plots of (a) linewidth vs electric-field strength, and (b) peak position vs electric-field strength for the QD shown in Fig. 1. The expressions used to fit the experimental data are shown (solid lines).

9.51 meV. Such a shallow confinement potential is consistent with the nature of the QDs in this sample, which are believed to be indium-rich islands on a damaged two-dimensional layer.

An external electric field F applied to a single QD is expected to produce a shift $\Delta E(F)$ in the exciton recombination energy which consists of both linear and quadratic terms. This shift arises from the QCSE and may be expressed as:

$$\Delta E(F) = \mu F - \alpha F^2, \quad (2)$$

where μ and α are, respectively, the components of the permanent dipole moment and the polarizability in the direction of the electric field.

Figure 2(b) shows a plot of peak position against field strength for the InGaN QD shown in Fig. 1. A fit was made to the plot using Eq. (2), yielding values of $\mu = -1.52 \times 10^{-28}$ C m for the lateral component of the permanent dipole moment and $\alpha = -2.32 \times 10^{-35}$ J V² m² for the lateral component of the polarizability.

The applied lateral electric field is applied perpendicular to the internal piezoelectric field (which generates a built-in QCSE shift within the InGaN QD). The application of the

external field produces an energy shift in the exciton recombination energy of up to 5.4 meV, much greater than the shifts reported in arsenide-based III/V QDs (up to 0.5 meV) and selenide-based II/VI QDs (up to 1.1 meV).⁷⁻⁹ This shows that the effect of the lateral electric field is not negated by the presence of the strong internal field in a perpendicular direction.

In conclusion, we have reported on the effect of externally applied lateral electric fields upon exciton recombination in a single InGaN QD. We observe a redshift in the exciton recombination energy of more than 5 meV, together with a reduction in the PL intensity. The field dependence, as expressed by $E(F) = \mu F - \alpha F^2$, is found to possess nonzero linear and quadratic contributions, indicating that the component of the permanent dipole moment in the lateral direction is not insignificant.

This work was supported by the Foresight LINK Award *Nanoelectronics at the Quantum Edge* (www.nanotech.org), by the EPSRC GR/R66029/01 and by Hitachi Cambridge Laboratory.

¹M. Sugisaki, H. W. Ren, S. V. Nair, K. Nishi, and Y. Masumoto, *Phys. Rev. B* **66**, 235309 (2002).

²S. Raymond, J. P. Reynolds, J. L. Merz, S. Fafard, Y. Feng, and S. Charbonneau, *Phys. Rev. B* **58**, R13415 (1998).

³S. A. Empedocles and M. G. Bawendi, *Science* **278**, 2114 (1997).

⁴J. H. Rice, J. W. Robinson, A. Jarjour, R. A. Taylor, R. A. Oliver, G. A. D. Briggs, M. J. Kappers, and C. J. Humphreys, *Appl. Phys. Lett.* **84**, 4110 (2004).

⁵Y. Nagamune, H. Watabe, M. Nishioka, and Y. Arakawa, *Appl. Phys. Lett.* **67**, 3257 (1995).

⁶K. Brunner, U. Bockelmann, G. Abstreiter, M. Walther, G. Böhm, G. Trankle, and G. Weimann, *Phys. Rev. Lett.* **69**, 3216 (1992).

⁷W. Heller, U. Bockelmann, and G. Abstreiter, *Phys. Rev. B* **57**, 6270 (1998).

⁸J. Seufert, M. Obert, M. Scheibner, N. A. Gippius, G. Bacher, A. Forchel, T. Passow, K. Leonardi, and D. Hommel, *Appl. Phys. Lett.* **79**, 1033 (2001).

⁹G. Bacher, H. Schömiß, J. Seufert, M. Rambach, A. Forchel, A. A. Maksimov, V. D. Kulakovskii, T. Passow, D. Hommel, C. R. Becker, and L. W. Molenkamp, *Phys. Status Solidi B* **229**, 415 (2002).

¹⁰R. A. Oliver, G. A. D. Briggs, M. J. Kappers, C. J. Humphreys, J. H. Rice, J. D. Smith, and R. A. Taylor, *Appl. Phys. Lett.* **83**, 755 (2003).

¹¹F. Widmann, B. Daudin, G. Feuillet, Y. Samson, J. L. Rouvière, and N. Pelekano, *J. Appl. Phys.* **83**, 7618 (1998).

¹²K. Tachibana, T. Someya, and Y. Arakawa, *Appl. Phys. Lett.* **74**, 383 (1998).

¹³C. Buchheim, A. Winzer, R. Goldhahn, G. Gobsch, O. Ambacher, A. Link, M. Eickhoff, and M. Stutzmann, *Thin Solid Films* **450**, 155 (2004).

¹⁴J. Seufert, M. Obert, M. Rambach, G. Bacher, A. Forchel, T. Passow, K. Leonardi, and D. Hommel, *Physica E (Amsterdam)* **13**, 147 (2002).

¹⁵B. Arnaudov, T. Paskova, B. Magnusson, E. Valcheva, B. Monemar, H. Lu, W. J. Schaff, H. Amano, and I. Akasaki, *Phys. Rev. B* **69**, 115216 (2004).