Giant permanent dipole moments of excitons in semiconductor nanostructures

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We have measured the vertical Stark effect of excitons confined to individual self-assembled ring-shaped quantum dots. We find that the excitons have very large permanent dipole moments corresponding to electron-hole separations up to 2.5 nm, comparable to the nanostructures’ physical height. We find a trend of both permanent dipole moment and polarizability on the emission energy, but a very strong correlation between the permanent dipole moment and the polarizability.

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Semiconductor quantum dots combine two contemporary themes: they are nanometre-sized and they can be self-assembled. They confine electrons and holes in all three directions, and this property is making them extremely attractive for photonics devices, not just for improved laser diodes,¹ but also for single photon sources² and detectors.³ There are many open questions concerning the strain and composition of quantum dots and how structural properties relate to electronic properties. Furthermore, the extent to which the electronic properties of quantum dots can be tailored is not yet known.

In the vertical direction, there are different confining potentials for electrons and holes leading to a spatial separation of the electron and hole wave functions, i.e., a permanent dipole moment. In addition, the exciton has a polarizability, the extent to which an applied electric field can pull the electron and hole apart, reducing the energy. Both parameters are important. The permanent dipole moment influences the excitonic oscillator strength and in addition, it is sensitive to the detailed structure of the dots. The polarizability determines the sensitivity of the exciton energy to an electric field. For applications of quantum dots as emitters and detectors, the polarizability should be large so that the energy can be tuned over a large range with modest electric fields. Conversely, for applications involving coherent control or high-resolution spectroscopy, the polarizability should be small because the Stark effect contributes to the line width, in particular of single dot emission, through the local electric fields generated by fluctuating charge.

InAs quantum dots in GaAs have an electron-hole separation of about 0.4 nm, with the holes localized at the apex of the dot, above the electron whose wave function is spread over the entire dot.⁴ The sign of this dipole moment has been interpreted as, first, evidence of a strong indium concentration gradient, with the apex indium-rich and the base indium-poor, and, second, of a truncation in the capped dots relative to uncapped dots,⁴ conclusions which have now been confirmed by structural studies.⁵,⁶ We present here measurements showing how we are able to tune the excitonic permanent dipole moment and polarizability of quantum dots by adjusting the self-assembly procedure.

Our experiments use quantum rings embedded in the insulator region of a metal-insulator-semiconductor heterostructure with which we have already demonstrated single electron charging of a single quantum ring.⁷ The quantum rings are situated 25 nm above an n⁺ region, and 150 nm below the surface. We evaporate a semi-transparent gate electrode on to the surface and apply a voltage Vg between gate and back contact. The quantum rings are grown by first depositing InAs on GaAs in the Stranski-Krastanow growth mode, producing lens-shaped quantum dots, 20 nm in diameter, 6 nm high, with a density in the 10⁹ cm⁻² range. We then deposit 1 nm of GaAs, after which we anneal at the growth temperature of 530 °C for about 30 s. During this annealing step, there is a significant movement of material laterally with a reduction in height to 2 nm. The astonishing feature is the appearance of a crater in the center of the dots, generating the ringlike structure.⁸,⁹ The radius at maximum height is ~18 nm. After the annealing step, the capping is completed by depositing more GaAs. The rings have a multi-peaked distribution, giving rise to a weak emission band at 1.27 eV and two strong emission bands at 1.30 and 1.34 eV in the ensemble photoluminescence. We have measured the photoluminescence (PL) of individual rings as a function of Vg at 4.2 K with a confocal microscope, using 300 nm sized holes in an opaque metal mask to resolve single rings.

Typical behavior is shown in Fig. 1. For Vg < −0.4 V, the PL is quenched because the electron tunneling time is smaller than the excitonic radiative lifetime.⁷ The single line for Vg ~ −0.3 V is the PL from neutral excitons (X⁰). At Vg = −0.24 V, the emission redshifts due to the formation of charged excitons (X⁻¹). We focus here on the Stark effect, the change in PL energy with vertical electric field. For each ring, we plot the PL energy as a function of Vg with the abrupt shifts in the PL energy subtracted. We find that the PL energy can always be fitted accurately to a quadratic function of Vg, exactly as expected for a polarizable dipole in an electric field. Two examples are shown in Fig. 2. We note that the Stark shift is not appreciably modified by any excess...
charge because the Stark shift of the Coulomb energies is small compared to the Stark shift of the electron and hole.

We describe an exciton in each ring with a permanent dipole moment $\mathbf{p}$ and a polarizability $\beta$, so that the PL energy $E_{\text{PL}}$ is given by $E_{\text{PL}} = E_0 + \mathbf{p} \cdot \mathbf{F} + \beta \mathbf{F}^2$, where $\mathbf{F}$ is the electric field. $\mathbf{F}$ is linearly related to the gate voltage $V_g$: $\mathbf{F} = -\frac{(V_g - V_0^0)}{d}$, where $V_0^0$ is the Schottky barrier potential and $d$ is the distance between the $n^+$ layer and the sample surface. We employ the sign convention that a positive $\mathbf{F}$ points from the wetting layer towards the sample surface. We have measured $V_g^0$ for our structure from the photocurrent induced by light with above GaAs-band gap energy. The signal changes sign for flat bands, determining $V_g^0 = 0.62$ V.

We take $d$ to be 175 nm, the target value from the growth. We have confirmed this value by measuring the capacitance between a known gate area and the back contact. Overall, we can determine $\mathbf{F}$ from $V_g$ to better than 10% accuracy.

We have determined $\mathbf{p}$ and $\beta$ for more than 20 individual rings. We express $\mathbf{p} = er$ where $e$ is the electronic charge and $r$ is the separation between the center-of-gravity of the electron and hole wave functions in the growth direction. In Fig. 3, we plot $r$ and $\beta$ against $E_0$, each point representing the result for one ring. Both $r$ and $\beta$ increase in magnitude with increasing $E_0$, but with large fluctuations from ring to ring. There are two surprising features. First, the permanent dipole moments are large. The average value is $\bar{r} = 1.2$ nm [standard deviation $\sigma(r) = 0.7$ nm], with some of the rings having dipole moments as large as $2.5$ nm. These values are both opposite in sign and much larger in magnitude than those previously reported (Ref. 12). Secondly, there is a clear linear relationship between $r$ and $\beta$, as shown in Fig. 4.

The permanent dipole moment is very sensitive to the details of the vertical electron and hole confining potentials. Calculations assuming pure InAs dots predict negative dipole moments, with the hole confined at the base of the dot, below the electron. Conversely, calculations assuming an indium-rich apex tend to predict positive dipole moments. This is because the indium-rich apex always has the largest uniaxial strain which has a powerful confining effect on the holes. The electrons are unaffected by the uniaxial strain, and they are lighter than the holes, enabling the electron wave function to delocalize over the dot. For uncapped dots grown in our chamber, we have very strong evidence from x-ray measurements for an indium-rich top and indium-poor base which in the light of all recent

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**FIG. 1.** A gray-scale plot of the photoluminescence (PL) of a single quantum ring versus gate bias $V_g$.

**FIG. 2.** The shift in PL energy versus electric field for two individual quantum rings, one weakly polarizable ($E_0 = 1.266$ eV, solid circles) and one highly polarizable ($E_0 = 1.363$ eV, open triangles). The solid curves are quadratic fits to the data.

**FIG. 3.** (a) The permanent dipole moment $p$, (b) the polarizability $\beta$, and (c) the energetic difference between the neutral and singly charged exciton $E_X$, plotted against $E_0$, the energy of the neutral exciton at zero electric field. Each point was recorded on an individual quantum ring. The symbols identify to which ensemble peak each ring belongs. Circle, triangle, square: ensemble PL peaked at 1.27, 1.30, 1.34 eV, respectively.
The Stark shift of all states is 

\[ E_F = -\frac{1}{2} m_e \omega_e^2 z^2 - eFz, \]

where \( \omega_e \) is the quantization energy, and \( m_e \) the effective mass. The Stark shift of all states is 

\[ A = -\frac{1}{2} \cdot 2 \cdot 2 = -2. \]

To interpret the results on the polarizability, we assume a parabolic confinement in the vertical direction \( z \), so that the Hamiltonian for an electron is 

\[ H = \frac{1}{2} m_e \omega_e^2 z^2 - eFz, \]

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from this that the indium profile of the rings is radically altered by our capping procedure. The polarizability increases with increasing emission energy which we interpret as a softening in the vertical potential. Finally, for a wide variety of rings, we uncover a linear relationship between the permanent dipole moment and the polarizability which we argue is a consequence of an almost ring-independent lateral extent. These results open up the possibility of tailoring the polarizability for particular applications.

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