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# Controlled generation of neutral, negatively-charged and positively-charged excitons in the same single quantum dot

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We report the controlled generation of neutral, negatively-charged and positively-charged excitons in the same single InAs quantum dot. The control parameters are a vertical electric field applied to a capacitor-like structure, in which the quantum dots are embedded, and optical pump power. The strong Coulomb blockade in quantum dots can be exploited to control the charge of excitons containing one hole, the neutral exciton, $X^0$, and singly negatively charged exciton, $X^{1-}$. We show here how this concept can be extended to excitons containing two holes, the biexciton, $2X^0$, and significantly the singly positively charged exciton, $X^{1+}$. We support all these assignments with a Coulomb blockade model. For all dots, the emission from the $X^{1-}$ is redshifted relative to the neutral exciton, but surprisingly we observe blueshifts as well as small redshifts for $X^{1+}$. *© 2005 American Institute of Physics*. [DOI: 10.1063/1.1937996]

Self-assembled InAs quantum dots are well suited for photonics applications. Their principal feature is a set of atom-like energy levels as a consequence of the three-dimensional confinement potential. This makes individual quantum dots attractive for single photon sources.[1] Additional applications in the areas of quantum optics and quantum information processing are underpinned by the long exciton coherence times in quantum dots.[2,3] It is clearly important to understand the level structure and Coulomb interactions within a single quantum dot. Controlling the charge offers a versatile tool to achieve this. Capacitance–voltage spectroscopy provides information on the electron–electron Coulomb interactions[4] and the energy shifts in photoluminescence (PL) are related to both electron–electron (e-e) and electron-hole (e-h) Coulomb interactions.[4,5] Additionally, charging is also of practical importance, since the singly charged excitons, $X^{1-}$ and $X^{1+}$, do not have a fine structure and are therefore well suited for single photon sources and due to the single excess spin left after recombination also for spin-based quantum information applications.

Generation of negatively-charged excitons has been reported, probing mainly the electron levels of the quantum dot.[4–6] The hole levels are likely to be much more sensitive to charging than the electron levels because the hole quantization energy is comparable to the Coulomb energies. $X^{1+}$ emission has been observed in power dependent experiments[7] and also from samples with a $p$-doped back contact,[8] leading to recent polarization-dependent measurements.[9] The motivation for the experiments reported here is to generate $X^{1+}$, $X^0$, and $X^{1-}$ in the same quantum dot in a controlled way. We have achieved this by developing structures which show perfect Coulomb blockade. At each bias, an exciton has sufficient time to enter the charge configuration with the minimum energy. The charge changes abruptly at particular bias voltages. As we show, there is a pronounced Coulomb blockade both for excitons with one hole and for excitons with two holes. We determine the dot parameters from the switching voltages of the $X^0$, $X^{1-}$, and $2X^0$ excitons and then predict the bias range in which the $X^{1+}$ exciton should exist. We find excellent agreement with the experimental results.

The InAs quantum dots in our experiment are embedded in a vertical tunneling structure with a tunnel barrier of thickness 25 nm between the dots and the $n^+$-doped back contact. The distance between the semitransparent NiCr top gate and the back contact is 175 nm and a GaAs/AlAs superlattice located 30 nm above the dots prevents carrier leakage to the gate. We use the emission of a VCSEL ($\lambda = 850$ nm) focused to a 1 $\mu$m spot to generate e-h pairs in the wetting layer of the quantum dots via photoexcitation, which then relax into the respective dot levels. At low power there is a sizable chance of creating an exciton, but negligible chance of creating a biexciton; at higher power there is also a sizable chance of creating a biexciton. By applying a voltage $V_g$ between the back contact and the top gate the quantum dot levels are shifted with respect to the back contact Fermi level allowing control over the exciton charge in the dot. Thus neutral or charged excitons are formed and by exploiting the pronounced excitonic Coulomb blockade we can control unambiguously the charging state of the dot with $V_g$ as the control parameter.[5] The PL generated as the exciton collapses is centered around 1.3 eV. It is collected with a confocal microscope and dispersed with a 0.5 m grating spectrometer onto a silicon charge coupled device array. The setup has a spectral resolution of 0.05 meV.

The upper part of Fig. 1 shows a contour plot of the main exciton lines of dot A at 4 K. The intense line appearing

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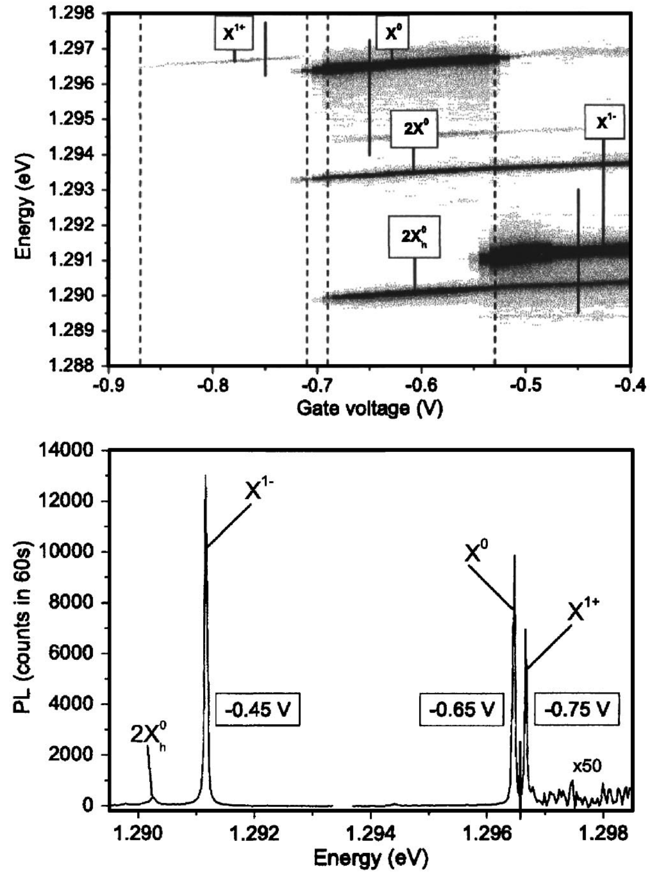


FIG. 1. The upper part shows a gray scale plot of PL from dot A at 4 K. White corresponds to 0 counts on the detector and black to 300. The main exciton lines have been labeled and the distinct charging events between them are indicated by dashed lines. $2X_h^0$ denotes a line related to $2X^0$ which we tentatively assign to a hot hole biexciton. The solid vertical lines mark the voltages for $X^{1-}$, $X^0$, and $X^{1+}$ at which PL spectra are shown in the lower part. They are labeled with the corresponding gate voltage and $X^{1+}$ has been enhanced by a factor of 50 for clarity.

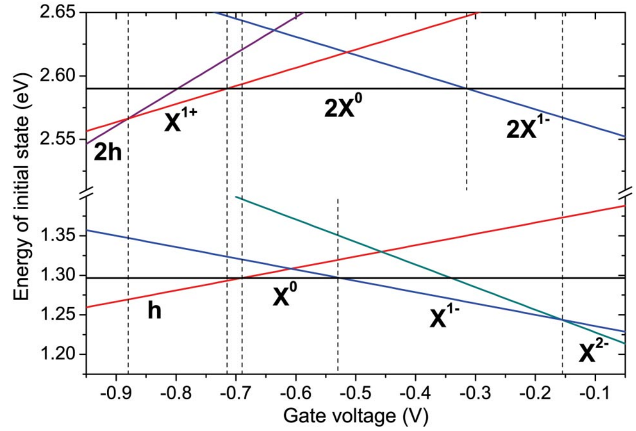


FIG. 2. (Color online) Gate voltage dependence of initial state configurations for the quantum dot A following our Coulomb blockade model. The energy of the vacuum state is taken to be zero. The lower group of lines corresponds to states with one hole, the upper group to states with two holes. Dashed lines indicate the voltages at which the charge of the ground states changes. The main energies are taken as $E_g = 1.328$ eV, $E_{ss}^{ee} = 25.8$ meV, $E_{ss}^{eh} = 31.5$ meV, $E_{sp}^{ee} = 23.5$ meV with a lever arm of seven following the notation of Ref. 4.

abruptly at −0.69 V and disappearing again at −0.53 V is the PL from $X^0$, the neutral exciton.[5] As the gate voltage is raised above −0.53 V the neutral exciton is replaced by $X^{1-}$, indicated by the sharp change in emission energy due to Coulomb interactions with the added electron. The large $V_g$ extent of $X^{1-}$ is due to the filled electronic s shell and large quantization energy of the dot.[5] At higher excitation powers more than one hole is occasionally present per dot, forming excitons such as the neutral biexciton, $2X^0$. This line can be identified from its superlinear dependence on excitation power and also from its large extent in gate voltage, an indicator for a filled s shell. At more negative gate voltages, $2X^0$ disappears and a new line appears. This line appears exactly when the $2X^0$ disappears, it too exists over a range of voltage before it fades out at large negative bias, and it is only visible when the pump intensity is high enough that there is a clear $2X^0$ signature. We have confirmed this result on several dots all of which show the same behavior. We attribute this line to emission from the $X^{1+}$ as it displays exactly the characteristics expected based on simple considerations of the Coulomb blockade. It is created from the $2X^0$ by removing an electron when the electrostatics favor this configuration, and the $X^{1+}$ itself is ionized to become the two-hole state at large and negative bias. In fact, the detailed behavior matches a model of the Coulomb blockade, as we show later. The lower part of Fig. 1 shows cross sections of $X^{1-}$, $X^0$, and $X^{1+}$ indicated by vertical lines in the contour plot. All exciton linewidths are resolution limited and within this limit neither $X^{1-}$ nor $X^{1+}$ shows any fine structure as expected. The PL emission of $X^{1+}$ is comparatively weak and about 20 times less intense than $2X^0$ for the given excitation power (compare Fig. 3).

To connect the behavior of $X^{1+}$ to the other excitons quantitatively we have developed a Coulomb blockade model. We assume strong confinement, treating the Coulomb interactions as perturbations to the single particle energies. We do not make any assumptions about the nature of the confining potential; instead we determine the e-e and e-h Coulomb energies from the excitonic Coulomb blockade. For example the energy of the neutral exciton state is the band gap energy $E_g$ reduced by the electron-hole Coulomb attraction $E_{eh}^{ss}$ with the electron and hole in their respective *s* states. We also assume the lever arm model[4] which implies a linear relationship between the gate voltage and the electrostatic potential at the quantum dot and we ignore the small vertical Stark effect. The calculation extends the approach of Ref. 4 to excitons with both one and two holes. The model gives the energy of a particular charge configuration as a function of gate voltage. Coulomb blockade arises because there are abrupt changes in the ground state charge as a function of $V_g$, as shown in Fig. 2 for the one-hole and for the two-hole excitons. In order to determine the dot parameters, we use the $V_g$ extents and emission energies of the $X^0$, $X^{1-}$, $X^{2-}$, and $2X^0$ lines for dot A. Based on this, we are able to make predictions on the behavior of $X^{1+}$. We find two main quantitative results. First, the $V_g$ extent of $X^{1+}$ should be exactly the same as the $V_g$ extent of $X^0$. Second, the transition from $X^{1+}$ to $2X^0$ occurs at a slightly more negative $V_g$ than the appearance of $X^0$, due primarily to the fact that the e-h Coulomb energy is larger than the e-e Coulomb energy.

We find excellent agreement of these predictions with the PL intensity plot in Fig. 3. Taking the voltages at which the lines reach half their maximum intensity as a measure of the $V_g$ extent, $X^{1+}$ switches on at about −0.71 V. It then fades at lower gate voltage, but can still be distinguished from the

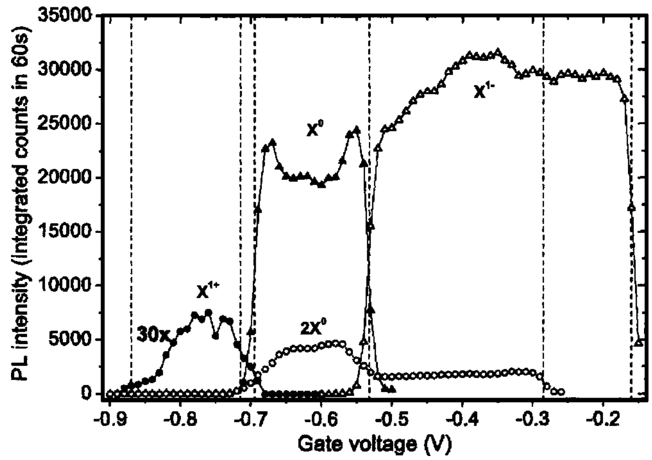


FIG. 3. PL intensities of exciton lines from dot A against gate voltage. There are clear cross-overs from $X^{1+}$ to $2X^0$ and from $X^0$ to $X^{1-}$. Note that $X^{1+}$ is about 20 times less intense than $2X^0$.

background out to −0.87 V. This gives a voltage extent of 160 mV. At higher voltages, $X^0$ exists between −0.53 and −0.69 V, an extent of 160 mV, exactly as for $X^{1+}$. Also, $X^{1+}$ makes a transition to the $2X^0$ at a more negative bias than the bias at which the $X^0$ appears, as predicted. Quantitatively, we predict that this voltage difference should be 24 mV for dot A; in practice, we measure 20 mV.

Two features of $X^{1+}$ warrant further discussion. First, the $X^{1+}$ emission line is about 20 times less intense than $2X^0$ and, unlike the $X^0$, does not switch off abruptly on the low gate voltage side but rather fades out. We propose that hole tunneling is responsible for this behavior. At large negative bias, the hole tunneling time decreases, ultimately becoming smaller than the radiative recombination time. This reduces the $X^{1+}$ emission intensity as the voltage is made more negative. We can support this assertion with measurements of the radiative decay time (not shown) which decreases from 0.3 to 0.1 ns in the region where the $X^{1+}$ decreases in intensity. The radiative recombination time for this dot is 0.8 ns, showing that tunneling dominates over radiative recombination at large negative bias. An interesting feature is that the effect of tunneling is more pronounced for $X^{1+}$ than for $2X^0$, presumably because the additional electron in the $2X^0$ provides additional binding for the holes.

The second feature is that we observe a wide range of shifts for $X^{1+}$ relative to $X^0$ ranging from blueshifts of up to 2.9 meV to small redshifts of 0.4 meV (see Table I). At first sight, this is surprising because on the same dots $X^{1-}$ is always redshifted relative to $X^0$ by about 5–6 meV. The result for $X^{1-}$ arises naturally from strong confinement and a hole wave function that is more localized than the electron wave function.[4] Applied to the $X^{1+}$, this picture would predict that the $X^{1+}$ is always blueshifted relative to the $X^0$, which is not the case in practice. We suggest that this discrepancy demonstrates that hole charging is not a small perturbation to the quantization, unlike the case of electron charging.

TABLE I. Energy of the neutral exciton for exemplary dots across the sample spectrum and corresponding shifts (in meV) in emission energy to $X^{1+}$, $2X^0$ and $X^{1-}$. Note that $X^{1+}$ can be blueshifted (dots A, B, C) as well as redshifted (dot D) relative to $X^0$.

| Energy | Dot A | Dot B | Dot C | Dot D |
|---|---|---|---|---|
| $X^0$ | 1.296 58 eV | 1.313 35 eV | 1.323 70 eV | 1.335 62 eV |
| $\Delta(X^0-X^{1+})$ | -0.41 | -2.38 | -2.86 | 0.36 |
| $\Delta(X^0-2X^0)$ | 3.04 | 1.48 | 1.41 | 3.24 |
| $\Delta(X^0-X^{1-})$ | 5.66 | 6.13 | 6.27 | 5.22 |

In conclusion, we have demonstrated the controlled generation of neutral, negatively-charged and positively-charged excitons in the same quantum dot by applying a voltage to a field-effect structure. We were able to understand quantitatively the voltage range in which the $X^{1+}$ line arises with a Coulomb blockade model. The $X^{1+}$ emission energies depart from a simple picture based on strong quantization, underlining the benefit of simultaneous electron and hole charging for a comprehensive picture of exciton behavior in a quantum dot.

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