Resonant transmission spectroscopy on the *p* to *p* transitions of a charge tunable InGaAs quantum dot

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We have performed resonant interband transmission spectroscopy on the transitions between the *s*-shells and the *p*-shells of a single charge tunable InGaAs quantum dot (QD). In contrast to the *s*-shell spectroscopy, investigating *p*-shell transitions allows the study of a QD charged with up to four electrons. The exciton charging state is clearly identified as a function of gate voltage ranges. In contrast to the *s*-shell, the *p*-shell electronic states show a strong tunnel coupling to the Fermi sea of the back contact. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2906900]

Resonant transmission laser spectroscopy on the exciton ground state of self-assembled quantum dots (QDs) has become a very powerful experimental tool.[1–3] It allows us, for instance, to investigate purely quantum optical effects such as power broadening and power induced transparency,[4] as well as the ac Stark effect of a two level system.[5] Furthermore, laser spectroscopy allows the measurement of the absorptive as well as the dispersive response of a QD,[6] the observation of the thermal occupation of the two Zeeman levels by an electron in a QD,[7] and the preparation of a single electron spin state with near unity fidelity.[8]

Most of the resonant single QD experiments have so far been performed on the exciton ground state; the transition between the *s*-shells (*s*-*s*) in the QD (see level scheme in Fig. 1 and the few experiments performed on the transitions between the *p*-shells (*p*-*p*) are limited to InAs QDs at laser wavelength above 1 μm.[1,9] The *s*-*s* exciton spectroscopy is only limited to neutral and singly charged QDs.[3] In contrast, resonant *p*-*p* exciton spectroscopy allows the investigation of QDs charged with up to five electrons with the prospect of revealing rich physics of few interacting electrons. In addition, resonant *p*-*p* transmission spectroscopy offers the possibility to study *p* to *s* intraband relaxation processes of electrons and holes[9] and its effects on nuclear spin polarization.[10] Furthermore, *p*-shell excitons were proposed as candidates for Kondo excitons, where the electron spin strongly interacts with an electron Fermi sea.[11] In particular, InGaAs QDs exhibit such a strong tunnel coupling.[12]

Photoluminescence excitation (PLE) is a standard tool to perform *p*-shell spectroscopy; however, while many PLE studies were performed on neutral and singly charged QDs,[10,13] very few papers are to be found on highly charged QDs with a clear charge identification. In this letter, we present a systematic mapping of *p*-shell exciton spectroscopy as a function of the QD charge.

Resonant *p*-*p* transmission spectroscopy was performed on a single InGaAs QD whose *s*-*s* transition has been shifted to around 950 nm by annealing to increase the tunnel coupling. The QD layer is embedded in a field effect structure to allow a tuning of the optical transitions by the Stark effect and a control of its charge. The structure consists of a *n*-doped back contact 25 nm below the QDs and a semitransparent metallic top gate 276 nm above the QD layer. An AlAs/GaAs blocking barrier was grown 10 nm above the QDs to prevent leakage currents. The back contact is grounded and a voltage applied to the top gate shifts the energy levels of the QDs with respect to the Fermi energy of the back contact. Due to the dimensions of the device, a change in gate voltage leads to a 12 times smaller change of the QD's energy. Electrons from the Fermi sea in the back contact tunnel into the QD and fill all states below the Fermi energy. Due to the strong Coulomb interaction, the number of electrons is exactly adjusted.[14]

The sample is mounted in a cryogenic confocal microscope with a spacial resolution of 1.2 μm to perform PL and transmission spectroscopy at 4 K. To study a single QD, a sample with a QD density of about 0.1 QD/μm^2 is chosen.

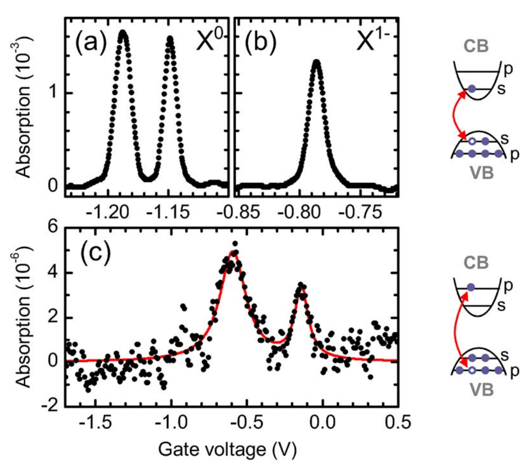

FIG. 1. (Color online) (a) *s*-*s* transmission spectrum of an uncharged QD as sketched on the right side. (b) spectrum of an *s*-*s* transition on the same QD charged with one electron. (c) *p*-*p* transitions as sketched on the lower right. The three resonances differ in charge. All data were taken under an intensity of 16 nW. The integration time per data point is 1 s for (a) and (b), and 20 s for (c).

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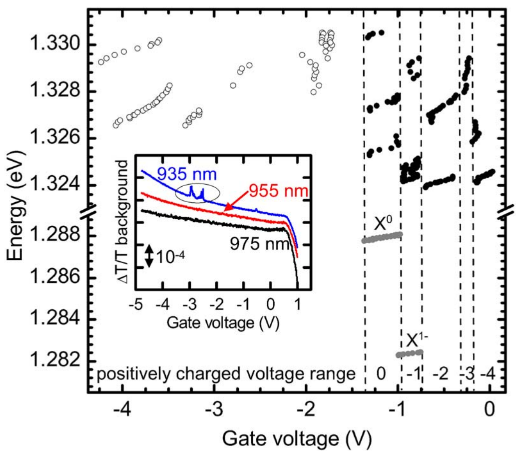

FIG. 2. (Color online) All observed resonance peak positions are plotted as a function of gate voltage. The charging events of the QD can be seen as jumps in energy and are marked by dashed lines. The charge states are listed. The inset shows the QD independent background signal for three laser wavelengths and the circle marks the QD resonances on top of this background.

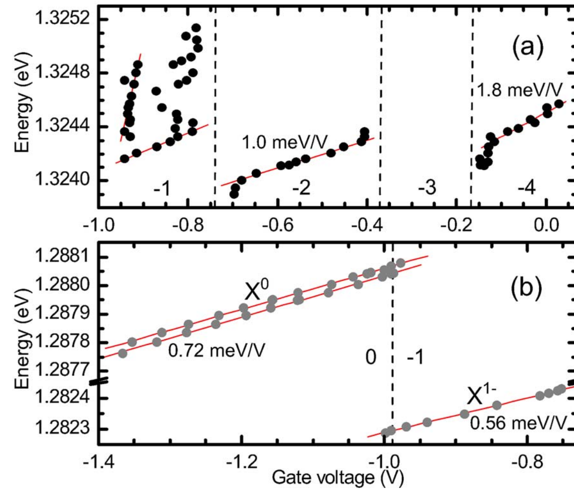

FIG. 3. (Color online) Zooms into Fig. 2 labeled by the charge state. (a) *p*-*p* transitions. The resonances for the QD charged with one electron show a rich structure. The two lines have slopes of 1.3 and 16.5 meV/V. The states charged with two and four electrons show clear plateaus but anticrossing with the Fermi energy at the charging voltages. (b) All observed *s*-*s* transitions are fitted with linear functions. The two charging states show a clear overlap and no departure from the linear behavior at the charging voltage.

This low density and intensive PL studies make sure that only one QD is in the focus of the objective and, hence, all observed transitions originate from the same QD.

To observe excitonic resonances, a wavelength tunable laser is set to a fixed wavelength and power, while the gate voltage is swept. The Stark effect shifts the QD's resonances through the sharp laser line (<1 MHz linewidth). A square wave signal is added to the gate voltage to use lock-in technique for noise reduction.[1,2] The choice of the laser wavelength is based on a preliminary PL and PLE mapping of the single dot spectra. This allows us, in particular, to identify the *s*-*s* transitions, their phonon replica, and the range of the *p*-*p* absorption photon energy.

In Fig. 1(a), the doublet of the *s*-*s*-X^0 transition on an empty QD is shown. The laser has an energy of 1.287 92 eV and, at the QD, an excitation power of 16 nW. The doublet arises from a symmetry breaking in the growth plain and the electron hole spin interaction.[2] The splitting is 39 mV corresponding to 28 μeV, as the Stark effect is 0.72 meV/V for this transition. The resonance lines of the neutral and singly charged excitons have linewidths of 5–10 μeV due to power broadening and show contrasts of $\sim 1.5 \times 10^{-3}$. The gate voltage extension of these two charge states can be seen in the lower part of Figs. 2 and 3(b). The value of the Stark effect needed to convert gate voltages in energies is obtained from linear fits to the individual plateaus, as shown in Fig. 3.

Figure 1(c) shows the *p*-*p* transitions at a laser energy of 1.324 15 eV. The *p*-*p* amplitudes are three orders of magnitude smaller than for *s*-*s* transitions, making the experiment challenging. Hence, integration time of 20 s instead of 1 s as for *s*-*s* had to be used to obtain a reasonable signal to noise ratio. The two resonances observed in this voltage trace show full widths at half maximum of 209 and 107 mV, corresponding to 209 and 193 μeV. The widths of the *p*-*p* resonances are, therefore, two orders of magnitude larger than for *s*-*s*. The sample has a wavelength dependent background which is independent of the presence of the QD, which is shown in the inset of Fig. 2 for the three laser wavelengths. QD *p*-*p* resonances are observed in the spectrum at 935 nm and are marked by a circle.

The gate voltage mapping of all observed resonances is presented in Fig. 2. The *s*-*s* transitions are depicted by gray dots in the lower half of the graph. The jump from X^0 to X^{1-} occurs when the lowest electron level is resonant with the Fermi energy of the back contact.[3] The 246 mV X^{1-} gate voltage plateau extension originates from the 21 meV charging energy for a second electron in the QD's *s*-shell.[3] The X^0 plateau expands over a voltage range of 372 mV caused by the electron hole Coulomb interaction of 31 meV. These charging energies are typical values for InGaAs QDs. The same charging voltages are observed for *p*-*p* transitions and are shown as black dots in the upper half of Fig. 2. The *p*-*p* transitions occur at energies about 40 meV higher than the *s*-*s*. In contrast to the *s*-*s* resonances, several lines are observed, even for the singly charged QD. Rich *p*-*p* spectra are expected from theoretical investigations for all charge states.[15] In addition to the two plateaus observed for the *s*-*s* spectroscopy, three more charge states are clearly identified by jumps in energy, marked in Fig. 2 by dashed lines and labeled by the charge of the QD. In analogy to the resonant *s*-shell spectroscopy and in contrast to PL,[3] *p*-shell resonant spectroscopy allows the measurement of the ground state charging energies but up to highly charged systems. Such pure electron charging has so far only been studied on self-assembled InGaAs QDs by capacitance spectroscopy on ensembles.[16] However, the InGaAs QD size distribution in ensembles smears out the features so that especially higher charge states cannot be resolved. We measure charging energies of 21, 34, and 11 meV for the second to fourth electrons.

As depicted in Fig. 2, we find resonances within the *p*-*p* exciton energy range at gate voltages more negative than −1.4 V for which the dot is expected to lose its electrons through exciton ionization. We estimate that the presence of trapped holes in the QDs lowers the electron energy below the Fermi level and prevents tunnel ionization.[3] In which case, we conclude that the observed states are positively charged excitons. We speculate that the holes feeding the dot are photocreated through the background absorption shown in the inset of Fig. 2. As seen in Fig. 1, the peak size of the positively charged *p*-*p* transitions is of the same order as the negative ones. For the 10^3 times stronger *s*-*s* transitions, no resonances are observed at gate voltages of $\leqslant -1.4$ V, and this within a noise level of 5×10^{-6} leads to the assumption that at this lower energies, considerable fewer free holes are photocreated.

For all *p*-*p* transitions, the Stark effect is significantly larger than that of the *s*-*s* transitions. For the whole range of measured gate voltage, it varies between 1.0 and 24 meV/V. This we logically relate to the weaker confinement of the *p* states along the growth direction. The weaker confinement has, furthermore, the effect of a stronger coupling to the Fermi sea which can be clearly identified by a strong nonlinearity in the voltage dependency close to the charging voltages. This effect can be interpreted as an anticrossing between the QD state and the Fermi energy of the back contact caused by strong tunnel coupling.[12] The stronger tunneling leads to a missing overlap of charge plateaus in the gate voltage in *p*-*p* spectroscopy, while in contrast, the *s*-*s* measurements process a strong overlap, as shown in Fig. 3(b). Figure 3(a) also shows a rich gate voltage dependency for the QD charged by one electron. Two lines are fitted with strongly different slopes of 1.3 and 16.5 meV/V. Also, for the QD with a single electron in the *p* shell (triply charged dot), a large slope of 24 meV/V is observed (see Fig. 2), giving rise to speculations about spin effects with the Fermi sea.

In conclusion, we have systematically mapped the *p*-*p* transmission resonant spectroscopy on a charge tunable QD. The measurement allows the observation of controlled QD charging with up to four electrons. Because of the weaker electron confinement in the *p*-shell, signatures of a strong coupling between the QD and the Fermi sea can be observed, while the *s*-*s* spectroscopy on the same QD serves as a well understood reference for weak coupling.

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