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Screening effects in InAs quantum-dot structures observed by photoluminescence and capacitance-voltage spectra

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We have performed photoluminescence spectroscopy as well as capacitance-voltage spectroscopy on an ensemble of self-assembled InAs quantum dots that are embedded in a field-effect-transistor structure. By investigating the charging spectra as a function of excitation power density, we are able to demonstrate a buildup of a transient positive charge in the heterostructure that leads to a screening of the electric field inside the structure. Moreover, by taking photoluminescence and capacitance spectra simultaneously, we can correlate the charging state of the dots with the interband transitions of *s*- and *p*-shell. We find that the observation of photoluminescence from higher orbital states in such field-effect-transistor structures is not only a consequence of Pauli-blocking but also of the accumulation of holes inside the structure. Also, we are able to determine the energy shift between the higher-charged excitonic states X^{2-} and X^{3-} to be $\Delta E = 2.6$ meV. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2112192]

In recent years, the Stranski–Krastanov growth method has been developed to become a versatile tool for the fabrication of semiconductor quantum dots.[1,2] These artificial structures, which confine charged carriers in all three dimensions, are attractive objects for studying many-particle interactions in fully quantized systems as well as promising candidates for novel optical and electronic applications. Many investigations have been performed to study the photoluminescence (PL) of self–assembled quantum dots, either as a function of the electron occupation number per dot[3,4] or of the excitation power.[5,6]

In this article we focus on the interband optical transitions, which we investigate in detail as a function of both, the number of electrons per dot and the excitation power. The electron occupation number can be controlled by the application of a suitable gate voltage and monitored by *in situ* capacitance spectroscopy. We will show that the electron occupation number, which is commonly assumed to be a function of gate voltage only, can be drastically affected by a change in the excitation power density. This might have an impact on quantum computing schemes that rely on Pauli-blocking effects.[7,8]

The samples used in this study have been grown by solid-source molecular beam epitaxy on a semi-insulating GaAs (100) substrate. The quantum dots (QD) are embedded in a field-effect-transistor (MISFET) structure.[2,3] First, a highly doped GaAs:Si back contact (BC) is deposited, followed by a 35 nm undoped GaAs layer which serves as a tunneling barrier. On top of this barrier, the InAs QDs are deposited, followed by 30 nm of undoped GaAs. To prevent tunneling between the QDs and the metallic top gate, a 172 nm thick AlAs/GaAs superlattice (SL) has been grown. The structure is capped with an 8 nm thick layer of undoped GaAs. Details on the growth procedure and sample preparation can be found elsewhere.[9,10]

By applying a gate voltage between the BC and the metallic top gate, the band structure [see Fig. 1(b)] can be tilted, so that the energy levels in the QD can be brought into resonance with the Fermi energy in the BC. In this case, electron tunneling between the QDs and the BC occurs, and an in-

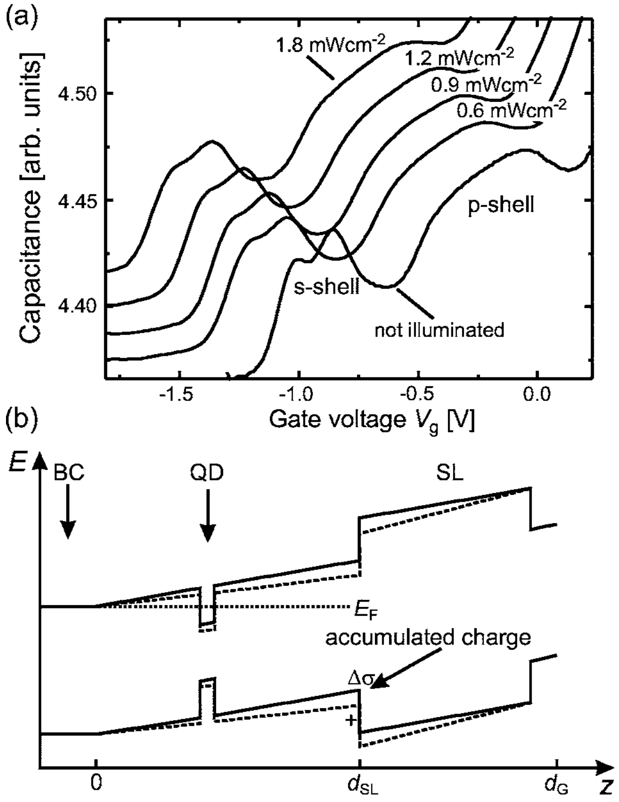

FIG. 1. (a) CV spectra taken at different excitation levels. A linear voltage offset of the CV spectra with increasing power density is observed. The spectra are offset vertically for clarity. (b) Schematic bandstructure without illumination (solid line) and under constant illumination (dashed).

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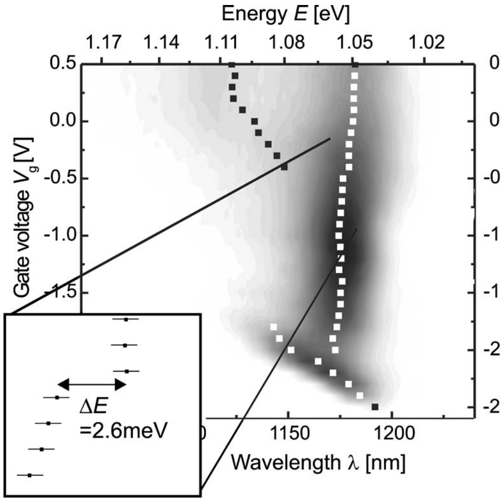

FIG. 2. PL spectra for gate voltages between $V_G=-2.6$ and 0.5 V at an excitation density of 12.6 mW cm^{-2}.

crease in the capacitance of the structure is observed.[2] Thus, the charging of single electrons per QD can be monitored.

The capacitance-voltage (CV) spectra are recorded in lock-in technique by superimposing a small AC voltage to a DC bias. PL spectra are recorded at $T=4.2$ K using a multimode fiber for both laser excitation and signal collection. As an excitation source a cw Ar$^+$ laser was used at $\lambda=514.5$ nm. The signal is dispersed in a Czerny–Turner monochromator ($f=550$ mm) and measured using an InGaAs detector.

First, we have measured the influence of laser excitation at different power densities on the CV charging spectra. The results are displayed in Fig. 1(a). The charging of the *s*- and *p*-shell can be clearly resolved for all excitation power densities. While the charging of the first electron for the unilluminated sample is observed at a gate voltage of $V_G=-1$ V, we find for higher excitation power densities the charging spectra to be offset by a negative voltage. This shift is found to be nearly linear with excitation density and the slope is 0.29 V/mW cm^{-2}.

We attribute the shift of the CV spectra in Fig. 1(a) to a transient buildup of positive charge at the GaAs/SL interface [see Fig. 1(b)], caused by a charge separation of the excited carriers in the built-in field of the structure.

The excitation laser generates electrons in the conduction band and holes in the valence band. Forced by the acting electrical field the electrons move in the direction of the back contact and the holes in the opposite way. In contrast to the electrons, which reach the back contact, the holes are trapped at the interface to the superlattice. The resulting positive sheet of charge will act upon the dot states like an effective positive internal potential, which has to be offset by an increased negative external gate bias. Using Gauss' law, we can directly relate the charge buildup at the GaAs/SL interface to the observed offset voltage ΔV_G. The charge density at the interface is given by

$$\Delta\sigma = \frac{\Delta V_G \cdot \varepsilon_0 \cdot \varepsilon_r}{d_G - d_{\mathrm{SL}}}, \tag{1}$$

where d_G is the distance BC–top-gate and d_{SL} the distance BC–SL. Using the above equation, we find a transient buildup of positive charge of 1.5×10^{11} cm^{-2} per 1 mW cm^{-2} excitation density.

In the next step, we have carried out ensemble PL measurements over a wide range of gate voltages and excitation power densities. For an excitation density of 12.6 mW cm^{-2}, the resulting spectra are plotted in Fig. 2. Between gate voltages of $V_G=-0.5$ and +0.5 V, two peaks are observed that can be associated to the interband transitions between the *s*-states (~1.05 eV) and the *p*-states (1.08–1.11 eV) in the QDs. In the gate voltage range between −0.5 and −1.7 V, the emission of the *p*−*p*-transition vanishes, and only the *s*−*s*-transition is observed. As the emission from the *p*−*p*-transition is suppressed, we observe a small blueshift $\Delta E=2.6$ meV in the energy of the *s*−*s*-transitions. To make this shift visible more clearly, we have magnified the region of interest in the inset in Fig. 2. The maxima in the intensity are plotted along with error bars as determined from the peak fitting procedure. We attribute this shift to the energy difference between the charged exciton states X^{2-} and X^{3-}. The value determined here is in good agreement with results from single dot spectroscopy[11] and values calculated theoretically.[12]

When the gate voltage is decreased beyond $V_G=-1.7$ V, a new peak arises with an energy of ~1.09 eV. While the intensity of this peak increases as V_G is reduced, the intensity of the *s*−*s*-transition is decreased, until it vanishes at about −2.0 V. At the same time, the energy of the emergent new peak shifts to lower energy, down to a value of ~1.03 eV for $V_G=-2.7$ V.

The fact that this new spectral feature increases at the cost of the *s*−*s*-transition strongly suggests that carriers in the QDs are involved in this recombination. However, as we do observe a strong electric-field-dependent shift, it is unreasonable to assume that both types of carriers are recombining from the QDs, as the quantum confined Stark effect would be too small to explain the large observed shift. Therefore, we conclude that a larger spatial carrier separation is responsible for this shift. As we have already demonstrated the presence of a hole accumulation layer at the GaAs/SL interface, we conclude that this spatially indirect recombination occurs between the *s*-shell QD electrons and holes accumulated at this interface. The observed shift of 47 meV for a gate voltage change of $\Delta V_G=500$ meV agrees well with a shift of 55 meV calculated by a lever arm argument for our sample geometry.[2]

Finally, we want to present our results for simultaneous measurements of PL and CV spectra. The analysis of these spectra allows us to correlate the interband transitions observed in PL to the charging state of the QDs as determined by CV. The results are displayed in Fig. 3. Here, the excitation density was as low as 2.2 mW cm^{-2}. It can be seen that the PL intensity of the *s*−*s*-transition has a maximum value, when the *s*-states are filled. The PL intensity of the *s*−*s*-transition drops, as the *p*-shell is filled with electrons, as seen in the CV-spectra. At the same time, the *p*−*p*-interband transition emerges in the PL signal. It is important to point out that the excitation density for these experiments was much too small for Pauli-blocking[13,14] to play a significant role, i. e., the *s*-states in the QDs were by far not saturated by the laser excitation. This was checked by measuring the PL for excitation densities up to 300 mW cm^{-2} (not shown here). However, we still do observe *p*−*p*-transitions even at these low excitation densities. Here, the combination of CV

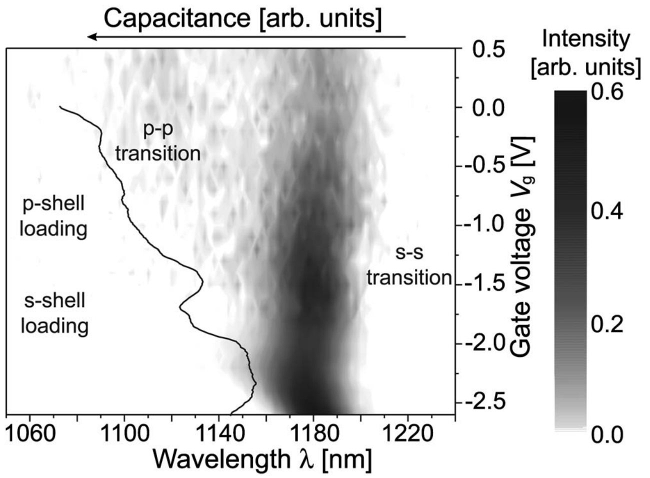

FIG. 3. Simultaneous measurement of PL intensity and CV in the range between V_G=−2.6 and 0.5 V. Optical interband transitions and electron charging of *s*- and *p*-states can be identified. The excitation power density was 2.2 mW cm^{-2}.

and PL proves to be a useful tool to identify the origin of the carriers participating in this process: As we see the *p*-shell loading when the $p-p$-transition emerges, we can safely conclude that the electrons are provided by tunneling from the BC. At the same time, we see the $s-s$-transition decrease, which is due to the fact that an increasing fraction of the holes captured by the QDs now recombine from the *p*-shell, thus reducing the $s-s$-recombination. Therefore, $p-p$-transitions can in this situation be observed without the presence of Pauli blocking.

In summary, we have shown that PL laser excitation in MISFET-QD structures causes a hole accumulation inside the structure, which leads to an offset in the charging spectra. For high applied gate voltages the holes in this layer can recombine with *s*-shell QD electrons. By correlating CV and PL spectra, we can identify the origin of carriers in optical transitions. This way, we could demonstrate that if electron tunneling in the QDs is possible, $p-p$ transitions can be observed even without Pauli blocking. This effect can be relevant for any structure that contains an interface where charge can be caught and has immediate consequences for any kind of PL experiments, where a MISFET structure is used to control the charging state of the QDs or to generate charged excitonic states. It therefore might be also relevant for recently proposed spin-based optical quantum computing via Pauli blocking in semiconductor quantum dots.[7,8]

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