Using a two-dimensional electron gas to study nonequilibrium tunneling dynamics and charge storage in self-assembled quantum dots

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We demonstrate a strong influence of charged self-assembled quantum dots (QD) on the conductance of a nearby two-dimensional electron gas (2DEG). A conductance measurement of the 2DEG allows us to probe the charge tunneling dynamics between the 2DEG and the QDs in nonequilibrium as well as close to equilibrium. Measurements of hysteresis curves with different sweep times and time-resolved conductance measurements enable us to unambiguously identify the transients as tunneling events between the 2DEG and QD states. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3175724]

Self-assembled quantum dots (QDs) are perfectly suited to study the electronic and optical properties in zero-dimensional semiconductor systems. In addition, they received much attention due to their great potential for new semiconductor devices, such as QD lasers, single photon sources[1] or future QD-based flash memories.[2] Thus not only for fundamental reasons, the knowledge of the internal electronic structure and charge carrier dynamics of QDs are of considerable importance. For the investigation of the electronic structure of self-assembled QDs, capacitance-voltage (*C*-*V*) spectroscopy has proven to be a valuable tool. Few-electron ground states and the corresponding charging energies can be probed with high accuracy in a static (near-equilibrium) capacitance measurement.[3–5] Furthermore, the charge carrier dynamics given by tunneling and thermally activated processes in large ensembles of self-assembled QDs can be observed in time-resolved capacitance measurements.[6,7] However, these capacitance measurements have experimental limitations in both time and spatial resolution. Therefore, studying few or even a single QD with long retention times is almost impossible using capacitance studies.

We introduce here a technique which enables to extend the experimental range regarding both tunneling dynamics and number of probed QDs. We show that the conductance of a two-dimensional electron gas (2DEG) can be used as an efficient detector to study the charge tunneling dynamics of nearby self-assembled InAs QDs with a time resolution ranging from microseconds to several tens of seconds. Furthermore this technique enables us to investigate nonequilibrium tunneling into excited dot states. The conceptual similarity of our sample structure to flash memories demonstrates the possibility to realize a QD memory device based on self-assembled QDs. Finally, the favorable scaling laws regarding the conductance of a 2DEG promises high-resolution single dot spectroscopy, which has already been successfully used to study lithographically patterned QDs.[8]

The investigated samples were grown in an inverted high electron mobility transistor structure with embedded self-assembled InAs QDs. They are based on the sample structure commonly used for *C*-*V* studies.[3,4] Instead of a highly doped 3D GaAs layer however, a 2DEG is used as a back contact.[5,9,10] The QDs are separated from the 2DEG by a tunneling barrier, which consists of a 10 nm $Al_{0.34}Ga_{0.66}As$ and a 20 nm GaAs layer. This results in charge tunneling times, which are orders of magnitude longer than those of previously investigated devices.[5,11,12] More details on the growth sequence can be found in Ref. 9. We have prepared Hall bar devices with a metallic top gate in order to control the charge state of the dots. The dot density of the sample is about 8.3×10^9 cm^{-2}, determined by atomic force microscopy studies of similarly grown dots on the sample surface. The gated electron channel area is 1.3×10^5 μm^2 which leads to about 1×10^7 probed QDs. Hall measurements yield a charge carrier density and a mobility of the 2DEG of about 7.4×10^{11} cm^{-2} and 9340 cm^2/V s, respectively. The conductance of the 2DEG is measured in a two-terminal geometry at a fixed source-drain voltage between $V_{SD}=30$ and 50 mV. All measurements are performed in a He cryostat at 4.2 K.

Figure 1(a) shows the *C*-*V* spectrum of the investigated sample. The observed maxima in the capacity can be directly linked to the individual electron states of the QDs.[4] The very weak coupling between the 2DEG and the QDs requires low-

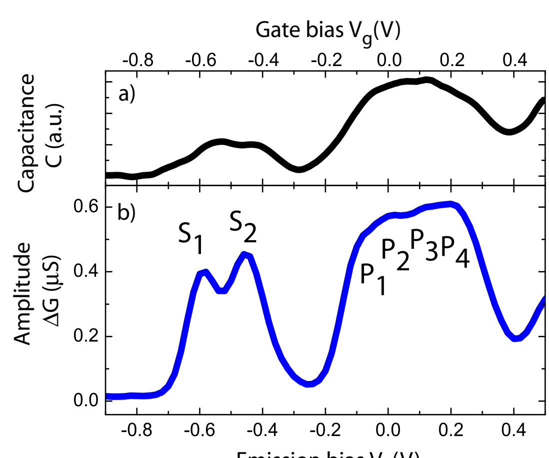

FIG. 1. (Color online) (a) *C*-*V* spectrum of the investigated sample. (b) The amplitude ΔG of the transients versus emission bias.

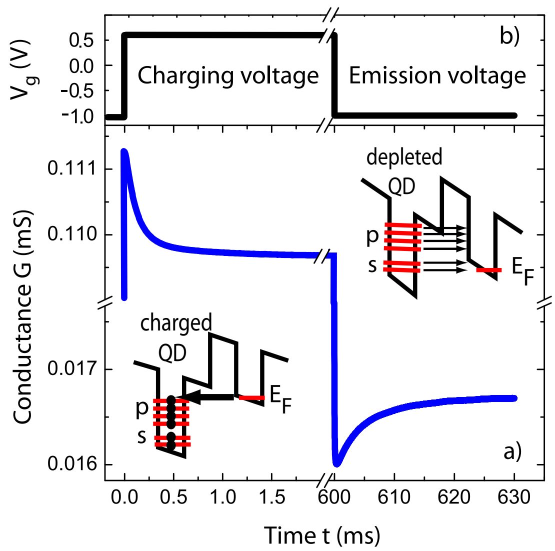

FIG. 2. (Color online) (a) Charging and emission transients from tunneling events between self-assembled QDs and a 2DEG measured via the conductance of the 2DEG. The schematic pictures illustrate the corresponding charging and emission process into and out of the QDs, respectively. (b) The corresponding applied gate bias V_g vs. time t.

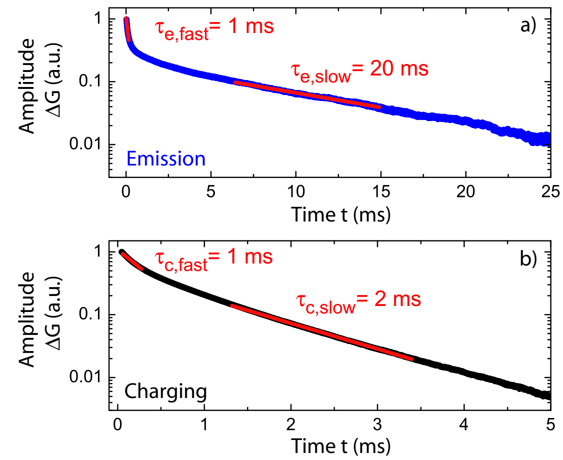

FIG. 3. (Color online) The normalized amplitude ΔG of the charging and emission transient from Fig. 2(a) on a semilogarithmic scale.

frequency modulation (23 Hz), which makes it difficult to obtain high-quality *C-V* spectra. However, a comparison with *C-V* studies from the literature[5] and the better resolved conductance measurements [Fig. 1(b), see discussion below] allows us to identify the double-peak structure around -0.5 V and the broad feature between -0.2 and 0.4 V with charging of the *s* and *p* shell, respectively. Accordingly, at a gate bias smaller than the charging voltage of the first *s*-state ($V_{g,s1} \sim -0.6$ V) the QDs are empty, and they are fully occupied (6 electrons per dot) at a gate bias larger than 0.4 V. Using frequency-dependent *C-V* spectroscopy,[12] the tunneling time of the first *s*-state is determined to be $\tau_{s1} \approx 6$ ms and the tunneling time of the *p*-states to be $\tau_p \approx 1.4$ ms. These relatively long charging and discharging times make it possible to directly study the dynamics of charge transfer between QD and 2DEG in the time domain.

Figure 2(a) shows the time-resolved conductance of the 2DEG when the gate voltage is changed abruptly [see Fig. 2(b)]. The time-resolved measurement starts with applying a charging voltage (V_c=0.6 V) to the gate electrode. As the Fermi-level E_F is now above the highest (*p*-) state of the QDs, electrons from the nearby 2DEG start to tunnel through the barrier into the dots (schematically depicted in the left inset in Fig. 2(a).

The charge carriers inside the QDs deplete the 2DEG. As a consequence, a decrease in the conductance can be observed which takes place in the first milliseconds, see left side of Fig. 2(a). At t=600 ms, an emission bias of V_e=-1 V is applied [Fig. 2(a)] which sets the Fermi-level E_F below the lowest (*s*-) states of the QDs (schematically depicted in the left inset) and tunneling from the QDs to the 2DEG takes place. Note that the time axis in the figure has been rescaled by a factor of 15 to account for the drastically different characteristic times for charging and emission.

To quantitatively evaluate the transient times, Fig. 3 shows the emission and charging transients of Fig. 2 on a semilogarithmic scale. The emission transient [Fig. 3(a)] shows a multiexponential decay with time-constants between $\tau_{e,\text{fast}}$=1 ms and $\tau_{e,\text{slow}}$=20 ms. Because tunneling is fastest out of high-energy states and slowest out of the low-energy states, we attribute the escape rate around $\tau_{e,\text{fast}}$=1 ms to tunneling out of *p*-state, however, the tunneling times of the *s*-states can be roughly limited up to $\tau_{e,\text{slow}}$=20 ms. This is in acceptable agreement with the frequency-dependent *C-V* measurements mentioned above with $\tau_p \approx 1.4$ ms and $\tau_{s1} \approx 6$ ms if the difficulties of estimating multiexponential decays are considered.[13] The charging process [Fig. 3(b)] also reflects a multiexponential transient. Surprisingly, however, only time-constants τ_c between 1 and 2 ms are observed (see corresponding linear fits in red). This discrepancy can be understood as a result of nonequilibrium tunneling processes as depicted by the insets in Fig. 2(a). During the emission process, the *s*-electrons have to penetrate a relatively high tunneling barrier [lowest arrows in Fig. 2(a), right]. During the charging process, on the other hand, because of the large positive bias the electrons are all injected into high-lying states with short tunneling times. The subsequent relaxation processes are known to be of the order of picosecond for electrons in self-assembled QDs.[14]

In order to study tunneling near-equilibrium and verify that the transients are caused by charge transfer between QD-states and 2DEG we use the *charge selective* operation.[15] The pulse amplitude is now small enough ($\Delta V = V_c - V_e$ =40 mV) to assure that not all 6 QD-states are affected but only an individual charging state is probed. The voltage V_e is scanned from -1 V (empty QDs) to 0.6 V (completely filled QDs). Figure 1(b) shows the transient amplitude $\Delta G = G(V_e,\ t=0\ \text{ms}) - G(V_e,\ t=600\ \text{ms})$ as a function of the V_e. Six individual charging peaks can be clearly identified and attributed to the charge occupation of the *s* and *p* shells of the QDs, in agreement with the results from standard *C-V* spectroscopy [see Fig. 1(a)]. However, the real-time measurement offers a much better resolution than the capacitance data. Moreover the clear charging signal in ΔG from the QD confirms that the transients in Fig. 2 and Fig. 3 are caused by electron tunneling from the different many-particle states in the self-assembled QDs. The present devices with very weakly coupled low-dimensional electron systems together with real-time conductance measurements enable us to decouple the applied gate bias from QD charge occupation.

The fact that nonequilibrium states can be prepared in the present sample makes them promising for charge storage as discussed in the following. Figure 4 shows the transfer

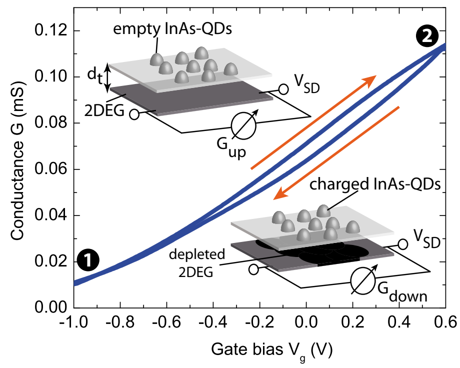

FIG. 4. (Color online) Conductance G of the 2DEG as a function of the gate bias V_g. The insets schematically depict the influence of the QDs on the nearby 2DEG.

characteristics of the sample, i.e., the conductance of the 2DEG versus the gate bias. The measurement cycle starts with a 200 ms long discharging pulse ($V_{\text{depl}}=-1$ V, point 1 in Fig. 4), which depletes the InAs QDs as discussed above (left inset in Fig. 4). A fast bias sweep ($\Delta t=2$ ms) from the depletion voltage upward to the filling voltage ($V_{\text{fill}}=0.6$ V) follows, faster than the average tunneling time between the QD-states and the 2DEG. As a consequence, QDs remain empty during the entire upward sweep and the 2DEG remains unaffected by the empty states of the QDs. Next, during a 200 ms long charging period at a gate bias V_{fill} of 0.6 V (point 2 in Fig. 4) the QDs become completely charged. Using Gauss' law to model the three layer system (gate, dot layer, 2DEG)[5] it can easily be shown that for every electron transferred into the QD layer roughly one electron will be depleted from the 2DEG, as schematically depicted in the right inset. The reduction in the charge carrier density lowers the conductance of the 2DEG, resulting in the observed hysteresis. The hysteresis decreases by increasing the sweep time and, hence, vanishes for sweep times longer than 200 ms, the longest charge carrier storage time in the QD ensemble (not shown here). Note that the influence of Coulomb scattering on the mobility due to the negatively charged dots is negligible.[5,10,16] The measured hysteresis opening $\Delta G/G$ in Fig. 4 for a gate bias of 0 V is about 10%. In comparison, the relative change in 2D carrier density between fully charged dots (6 electrons per dot) and empty dots amounts to $\Delta n/n \approx 7\%$ at $V_g=0$ V. Using an approximately constant mobility[5] leads to $\Delta G/G \approx 7\%$, in good agreement with the measured value. Thus, we are able to switch between two different QD charge occupation levels (completely full and empty QDs) for the same applied gate bias. This further supports the conclusion that the observed hysteresis is indeed given by the different QD charging states. Other groups have observed a similar hysteresis effect using laterally patterned electron channels[17,18] or optical excitation to discharge the QDs.[19] Some of the hysteresis effects could related to charge storage in deep-levels.[20]

So far, the electron and hole dynamic of self-assembled QDs have mostly been studied in time-resolved capacitance measurements.[2,15] For both high density storage application and high resolution spectroscopy, single QD measurements are the ultimate goal. Capacitance measurements, however, have a serious disadvantage because the capacitance scales with the area of the investigated sample, which is well below μm^2 for single dot studies. The presented conductance measurements of a nearby 2DEG could overcome this drawback and enable to probe single charge carrier dynamics. The conductance of a 2DEG is only determined by its lateral geometry (width/length). Therefore, the observed signal $\Delta G=G \times N_{\text{QD}}/N_{\text{2DEG}}$ is constant for a given QD density N_{QD} and carrier density in the 2DEG N_{2DEG} and, hence, independent on the sample size and number of QDs involved. This estimation supports our conclusion that this technique could be used to study the carrier dynamics of single self-assembled QDs as successfully shown before for lithographically defined QDs. This would make it possible to apply techniques that have been highly successful for single lithographic dots[8] also for studying self-assembled QDs.

To summarize, we have reported on a time-resolved conductance measurement technique with greatly enhanced experimental possibilities to investigate very weakly coupled low-dimensional electron systems. We have demonstrated that a 2DEG can act as a sensitive detector and could identify clearly the QD electron tunneling in the measured transients.

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