Charging dynamics in vertically aligned InAs quantum dots

R.J. Luyken[a], A. Lorke[b], G. Medeiros Ribeiro[c], P.M. Petroff[c]

[a] Infineon technologies AG, Corporate Research , Otto-Hahn-Ring 6,
81730 München

[b] Sektion Physik, LMU München, Geschwister Scholl Platz 1 80539 München

[c] Materials Department and QUEST, University of California, Santa Barbara,
California 93106

Using frequency-dependent capacitance spectroscopy we investigate the dynamics of tunneling from a three-dimensional back contact into ensembles of self assembled InAs quantum dots. An equivalent RC-circuit is derived from the balance of charge and allows us to determine the charging time for each state of the dots. This method is applied to investigate the charging dynamics of samples with double layers of vertically aligned InAs quantum dots. In these structures the interplay between Coulomb blockade effects and sequential tunneling can be seen: The charging time for the dots in the second layer is either reduced by sequential tunneling through states of the first dot layer or enhanced by Coulomb blockade effects.

Recently, self-assembled strained islands have attracted particular interest as they provide for well-defined, nanometer-size quantum dots with sizes in the 10 nm range[1]. These systems are of great interest, not only for studying the basic properties of man-made „artificial atoms“, but also because of possible device applications. Using capacitance and far-infrared spectroscopy, the many-particle ground states as well as the excitations of the dots have been previously investigated in detail[2-4] as well as the optical properties of these promising candidates for improved laser design[5].

For possible device applications of InAs dots in the future it is crucial to understand the charging dynamics of these structures. Here we investigate the charging dynamics from a three-dimensional back contact into ensembles of self-assembled InAs quantum dots by frequency-dependent capacitance spectroscopy. Previously the influence of Coulomb blockade, magnetic field and temperature has been described[6]. Here we focus on the derivation of an equivalent RC-circuit for the determination of the averaged charging time and the special effects that take place in samples with two layers of vertically aligned InAs quantum dots. It is shown that the interplay between Coulomb blockade effects and sequential tunneling has a strong effect on the charging characteristics of the dots in the second layer.

The samples are grown by molecular beam epitaxy, using the Stranski-Krastanov growth to create the self-assembled InAs quantum dots. The dots are embedded into a suitably designed metal-insulator-semiconductor-field-effect-transistor (MISFET) type GaAs/AlGaAs heterostructure as described in Ref. 3, 7. Fig. 1 shows a sketch of the conduction band edge of a single layer sample (1a) and a double layer sample (1b). In the double layer samples the dots in the second layer are aligned along the growth direction to the dots of the first layer; It is assumed that the strain field of the first dot acts as a nucleus for the growth of the second dot. Except for the second dot layer the samples are identical to the single layer samples. Using the simple lever arm argument a voltage dV applied between the gate and the back contact can be converted approximately into an energy shift of the dots $\Delta E = e\Delta V d_1 /(d_1 + d_2)$. The samples are provided with Ohmic contacts and a semi-transparent gate. The gate area of the investigated samples are between 0.1 mm^2 and 2 mm^2; thus with typical dot densities of 10^{10}/cm^2, ensembles with typically $1*10^7$ – $2*10^8$ dots are probed. The samples are mounted in a liquid He cryostat and the capacitance–voltage (CV) spectra are measured using a dual-phase lock-in amplifier.

As described in Ref. (2,3) the lowest and second-lowest single electron states of the InAs quantum dots are doubly and fourfold degenerate, respectively. For the many particle states this degeneracy is lifted by electron-electron interactions, so that groups of charging peaks appear, which, in analogy to atomic physics, are commonly labeled s- and p-shell.

Fig. 2 shows CV traces of a single layer sample with $d_1 = 40nm$ and $d_2 = 200nm$ at frequencies from 2 kHz up to 400 kHz at T= 4.2k. The background capacitance between the gate and the back contact increases linearly with and has been subtracted. At first glance it can already be seen that increasing the frequency affects the different states differently: the s-states are strongly, the p-states more weakly and the wetting-layer is not affected at all in this frequency regime. The limitation is here the oscillator frequency of the lock-ln amplifier, but not the physical properties of the device: On samples with larger tunneling barrier both the p-states and the wetting layer are (at sufficiently high frequencies) affected in the same way than here the s-states. The different tunneling times can be explained qualitatively by the fact that an increase in gate voltage results in a decrease of the tunneling barrier heights. A more qualitative analysis is given in Ref. 6.

Derivation of a RC-equivalent circuit from the balance of charge

From the balance of charge within the MISFET structure we derive in the following the RC equivalent circuit picted out in Fig. 3b

Assuming an applied Voltage of the form

$$U = U_0 + \Delta U e^{i\omega t} \tag{1}$$

results in an linear response of the area charge density both in the backcontact σ_B and in the quantum dot layer σ_D

$$\sigma_B = \sigma_{B0} + \Delta\sigma_{B0} e^{i(wt+\beta)}\,; \qquad \sigma_D = \sigma_{D0} + \Delta\sigma_{D0} e^{i(wt+\delta)} \tag{2a}$$

$$\Delta\sigma_B = \Delta\sigma_{B0} e^{i(wt+\beta)} \qquad \Delta\sigma_D = \Delta\sigma_{D0} e^{i(wt+\delta)} \tag{2b}$$

where β and δ are the phase shifts of the area charge density with respect to the applied signal. The current j(t) which flows from the external circuit into the structure is proportional to the measured capacitance signal and is given by

$$j(t) = \frac{d}{dt}(\sigma_B + \sigma_D) \tag{3}$$

From the definition of the capacitance –here related to an area charge density σ - $dU = d\sigma / C$ the deviation dU from the equilibrium U_0 can be written as

$$dU = d\sigma_B \cdot \frac{d_1 + d_2}{\varepsilon_0 \varepsilon} + d\sigma_D \frac{d_2}{\varepsilon_0 \varepsilon} \tag{4a}$$

$$\frac{dU}{dt} = i\omega \Delta U e^{i\omega t} = \frac{d_1 + d_2}{\varepsilon_0 \varepsilon} i\omega \Delta\sigma_{B0} e^{i(\omega t+\beta)} + \frac{d_2}{\varepsilon_0 \varepsilon} i\omega \Delta\sigma_{D0} e^{i(\omega t+\delta)} \tag{4b}$$

It is to note that the frontgate is chosen as the reference point for the voltage deviation (s. Fig. 3a). The charge transfer between the quantum dot layer and the back contact is described by a decay-equation with a tunneling time τ

$$\frac{d\sigma_D}{dt} = -\frac{eD\Delta\mu}{\tau} = \left(\frac{\Delta\sigma_D - eD\Delta\Phi}{\tau}\right) \tag{5a}$$

$$i\omega\tau e^{i(\omega t+\delta)}\Delta\sigma_{D0} = -\frac{1}{\tau}\left(\Delta\sigma_{D0}e^{i(\omega t+\delta)} - e^2 D\frac{d_1}{\varepsilon_0\varepsilon}\Delta\sigma_{B0}\right)e^{i(\omega t+\beta)} \tag{5b}$$

where D is the density of states in the quantum dot layer. The temporary evolution of the charge density σ_D in the quantum dot layer is proportional to the difference in the electrochemical Potential[8] $\Delta\mu$ between the back contact and the quantum dot layer. The electrochemical potential contains both a contribution from $\Delta\Phi$, the potential drop between the backcontact and quantum dot layer and from $\Delta\sigma_D$, the deviation from the charge equilibrium in the quantum dot

Solving eq. 5b for $\Delta\sigma_D e^{i\delta}$ and substitution in eq. 4a results in an equation of ΔU dependent on $\Delta\sigma_B e^{i\beta}$. Substitution in eq. 3 leads to in

$$j(t) = i\omega e^{i\omega t}\left(\Delta\sigma_B e^{i\beta} + \Delta\sigma_D e^{i\delta}\right) = i\omega e^{i\omega t}\Delta\sigma_B e^{i\beta}\left(1 + \frac{d_1}{\varepsilon_0\varepsilon}\cdot\frac{e^2 D}{1+i\omega t}\right) \tag{6}$$

with a further substitution of eq. 4 the current can be expressed through the sample parameters d_1, d_2, D and τ

$$j(t) = i\omega e^{i\omega t}\Delta U\left(\frac{\dfrac{\varepsilon_0\varepsilon}{d_1} + \dfrac{e^2 D}{1+i\omega\tau}}{1 + \dfrac{d_2}{d_1} + \dfrac{d_2}{\varepsilon_0\varepsilon}\cdot\dfrac{e^2 D}{1+i\omega\tau}}\right) = i\omega e^{i\omega t}\Delta U C_2\left(\frac{C_1 + \dfrac{C_q}{1+i\omega\tau}}{C_1 + C_2 + \dfrac{C_q}{1+i\omega\tau}}\right) \tag{7}$$

$$\text{with } C_1 = \frac{\varepsilon_0\varepsilon}{d_1};\quad C_2 = \frac{\varepsilon_0\varepsilon}{d_2};\quad C_q = e^2 D \tag{8}$$

With $\tau = RC_q$ the RC- equivalent circuit depicted in Fig. 3b results in the same current as described in eq. 7. The imaginary part of eq. 7 which is measured in the capacitance spectroscopy and noted here as $C(\omega)$ can be (after subtraction of a background capacitance) brought into the form of a Lorentz-function

$$\frac{\Delta C(\omega)}{\Delta C(\omega \to 0)} = \frac{C(\omega) - C(\omega \to \infty)}{\Delta C(\omega \to 0) - C(\omega \to \infty)} = \frac{1}{1 + (\omega\tau\delta)^2} \tag{9}$$

with $\delta = \dfrac{C_1 + C_2}{C_1 + C_2 + C_q}$ (10)

C_1, C_2, C_q can be determined from the sample parameters. Thus, using eq. 9 the averaged charging time τ can be derived from fitting the measured capacitances to eq. 9. For the data of Fig. 2 this is shown in Fig. 3c and the charging times can be determined to $\tau_{s1} \approx 500\mu s$ for the s$_1$-state and $\tau_{s2} \approx 270\mu s$ for the s$_2$ state. It is to note that the RC-circuit in Fig. 3b differs by the additional "quantum capacitance" C_q from the commonly used RC circuit (Ref. 7 and references therein). Therefore the method described above could also be useful to include quantum effects into the charging dynamics of other structures.

Charging effects in vertically aligned InAs dots

In the experiments with single layer samples as in Fig. 1a and Fig. 2 the dots are fairly dilute, so that they can to good approximation be treated as non-interacting. For the investigations of coupled dots, samples with two layers of vertically aligned InAs quantum dots have been used[9]. Coupled dots are relevant both from a viewpoint of building artificial molecules and from possible implementation of quantum dots in future devices. Previously it has been shown that a strong electrostatic dot-dot interaction reveals itself in distinct shifts of the many particle ground state[9]. In the following the particularities that take place in the charging dynamics of samples with two layers of quantum dots are discussed. (sketch of the conduction band edge of the investigated double layer samples is shown in fig. 1b, $d_{11} = 25nm$, $d_2 = 150nm$). In the investigated samples the dot-dot distance d_{dd} was either 10nm or 20nm. According to d_{dd} the samples are in the following labeled as 10nm sample and 20nm sample. Fig. 4 shows the CV spectra of the 10 nm samples recorded at different frequencies. Up to Vg=-0.2V, the trace is almost identical to a trace of a single layer sample. Around Vg=0.1 V and Vg=0.3 clear peaks appear which can be identified as the s- and p-shell of the second dot layer. For the s-shell a frequency dependence is observed and applying the method described before we determine the charging time for the s-shell to $\tau = 25ms$. In this structure the distance from the back contact to the dots of the second layer is 45 nm. On a single layer sample with this distance to the dots we observe a charging time of $\tau = 1ms$. Thus in the double layer sample the charging time is enhanced by a factor of 25 compared to the single layer sample. This effect is attributed to the Coulomb repulsion from the first dot layer which is filled with 6 electrons for each dot. A different picture however is observed in the sample with 10nm dot-dot distance. The corresponding CV-spectra is shown in Fig. 5. Here deviations from the spectra of a single layer sample already occur within the charging of the p-shell, as the charging of the second dots occurs after the p-level of the first is filled with 2 electrons. It can also seen from Fig. 5 that increasing the frequency does not affect the spectra. Up to 1 MHz, the upper limit of the lock-in amplifier that was used, no suppression of the charging signal as in Fig. 2 or Fig. 4 was observed. On the other hand, in a single layer sample with the same distance to the dots (35 nm) we observe a suppression of the charging signal starting from 20 kHz. Thus, in the double layer sample even as there is also the Coulomb-repulsion from the first dot layer the charging time is reduced compared to the single layer sample. This effect is attributed to sequential tunneling: In the 10 nm sample there are states in the first layer which act as a bridge for charging of the second layer. In the 20 nm sample, however the s-state energies of the second dot lie in the gap between the p- and d-shell of the first dot and thus there are no bridge states.

In conclusion we have demonstrated that from the balance of charge an equivalent RC-circuit can be derived which includes a quantum capacitance. On samples with vertically aligned InAs dots we observe that the charging dynamics is dominated by the interplay between Coulomb-repulsion and sequential tunneling.

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Figure captions

Fig. 1

Sketch of the conduction band edge along the growth direction of the investigated samples along the growth direction with respect to the Fermi level E_F

a) single layer sample

b) double layer sample .

Fig. 2

Capacitance-voltage (CV) traces of a single layer sample with $d_1 = 40nm$ and $d_2 = 200nm$ recorded at frequencies 2.3 kHz, 8.3 kHz, 13.8 kHz, 20.1 kHz, 33 kHz, 43 kHz, 101 kHz, 200 kHz and 400 kHz (T=4.2 K). The background capacitance between the gate and the back contact has been subtracted.

Fig. 3

a) Schematic to show the charge distribution within the MISFET structure
b) Equivalent RC circuit derived from the balance of charge (see text). From the commonly used RC circuit it differs by the "quantum capacitance" C_q.
c) Capacitance amplitude of both s_1 and s_2 state plotted against frequency and fitted according to Eq. 2 in the text.

Fig. 4

Capacitance-voltage (CV) traces of a double layer sample with $d_{11} = 25nm$, $d_{dd} = 20nm$, $d_2 = 150nm$ recorded at frequencies 21 Hz, 41 Hz, 81Hz and 231 Hz. (T=4.2 K).

Fig 5

Capacitance-voltage (CV) traces of a double layer sample with $d_{11} = 25nm$, $d_{dd} = 10nm$, $d_2 = 150nm$ recorded at frequencies 713 Hz and 787 kHz. (T=4.2 K).