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Fine structure in the spectrum of the few-electron ground states of self-assembled quantum dots

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Abstract

We study the ground states of self-assembled InAs quantum dots with high-resolution capacitance spectroscopy. For samples with a relatively large gate area, and therefore large number of dots, we observe essentially single-electron charging into the lowest-energy states of the dots. A complete picture of the evolution of the ground states with electron occupation and magnetic field is established. For smaller samples, an additional fine structure is seen in the capacitance spectra. Its relative amplitude increases with decreasing gate area of the device. The typical energy spacing between the additional maxima is about 5 meV. It is discussed in terms of mesoscopic fluctuations in the capacitance device. For samples containing very few dots the spectra consist of sharp charging peaks, some of which are separated by the typical energy of the fine structure. © 1998 Elsevier Science B.V. All rights reserved.

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Self-assembled quantum dots are an ideal tool to study the energy states of zero-dimensional electron systems [1–3]. In these systems, the spatial quantization caused by the in-plane confinement is so strong that the single-particle energies exceed the Coulomb charging energies [3]. Here we study how the capacitance spectra which reflect the electronic ground states change with decreasing sample areas.

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The samples are grown by molecular beam epitaxy [4,5]. The InAs dots are embedded into a MIS-type heterostructure as described in Refs. [3,5,6]. From atomic-force micrographs we estimate the dots to be about 20 nm in diameter and 7 nm in height [4]. The dot density is in the range of $10^{10}\,\mathrm{cm}^{-2}$ [4,5]. In these capacitance devices the area A of the metal-gate electrode on the crystal surface is a rough measure of the dot number in the ensembles under investigation. The number of electrons per dot can be tuned with the gate voltage V_g applied between gate and back contact.

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A high-resolution capacitance bridge is employed to measure the capacitance–voltage (CV) characteristic of the dot arrays [7,8]. The signal at the balance point is detected with a phase-sensitive amplifier via an on-chip impedance transformer. More details of the measurement set-up can be found in Refs. [8,9]. With this technique we can resolve capacitance changes down to aF.

An increased capacitance signal with respect to the background reflects electron tunneling from the back contact into the dots. In a simplified model, this occurs at the gate voltage at which the total energy for N and $N+1$ electrons in the dots is the same and the number of electrons per dot can change [10,11]. Dividing the gate voltage differences by the lever arm d_{tot}/d_t, where d_{tot} is the distance between back contact and front gate and d_t is the thickness of the tunneling barrier between back contact and dot layer, gives an estimate for the corresponding energy differences [3].

Fig. 1 shows the CV traces of a sample with a relatively large gate area, $A = 89\,\mu m^2$, at different magnetic fields applied in the growth direction. We

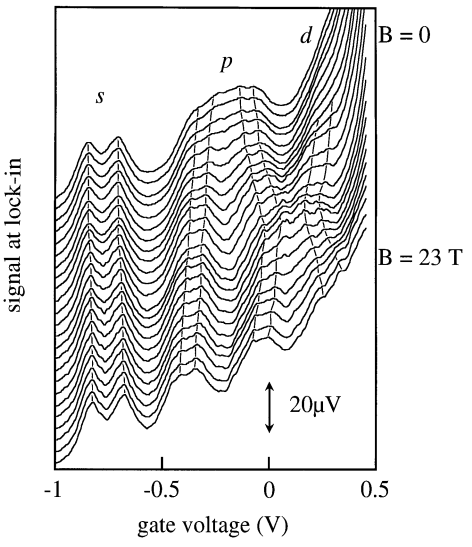

Fig. 1. Differential capacitance for a sample with $A = 89\,\mu m^2$ at different magnetic fields applied in the growth direction. From top to bottom the field is increased in steps of 1 T from $B = 0$ to 23 T. The conversion factor from voltage signals at the lock-in input to capacitance changes in this sample is roughly 50 aF/μV.

concentrate first on the trace at $B = 0$. At low gate voltage, $V_g < -1$ V, the signal is determined by the parallel-plate capacitor formed by the backcontact and front gate. The increase of the signal at positive gate voltage, $V_g > 0.2$ V, reflects the charging of the two-dimensional wetting layer from which the dots are initially formed. In between, the charging of the dots with single electrons can be seen as distinct peaks. A double structure around $V_g = -0.8$ V and four maxima in the broad feature around $V_g = -0.25$ V are observed.

Due to the disk-like shape of the dots we assume the lateral quantization energy to be separable from the much larger quantization energy in the growth direction. In the Fock model [12], i.e. a single-particle model which assumes a two-dimensional parabolic confinement, the first energy state is twofold and the second energy state fourfold degenerate. We approximate the action of the electron–electron interaction by adding a Coulomb blockade term to the Fock states which lifts the degeneracy of the states. According to the angular quantum number we describe the lowest and second lowest-energy states as s- and p-like, respectively.

We interpret the double structure at $V_g = -0.8$ V as Coulomb blockade in the charging of the s-(like) shell. From the gate voltage difference $V_g^{12} = 132$ mV between these peaks an s-shell Coulomb charging energy of 21.5 meV and a lateral quantization energy $\hbar\omega_0 = 44$ meV can be derived [9]. From the integral of the CV characteristic over the s-shell the dot number under the gate can be estimated to be about 3500. The Coulomb blockade of the p-shell is observed as well. As can be seen in Fig. 1, the Coulomb blockade depends on the number of electrons in the dots. We measure $V_g^{34} = 61$ mV and $V_g^{56} = 67$ mV. This dependence of the Coulomb charging energy on the number of occupied states can be explained by the effective dot size which is larger for the p-electrons than for the s-electrons. The large value $V_g^{45} = 128$ mV indicates the importance of the exchange interaction due to spin [9,13].

All measurements discussed here were performed at $T = 4.2$ K with a modulation amplitude of 4 mV. It should be mentioned at this point that we are able to resolve the Coulomb blockade of the s-shell even at $T = 77$ K.

The classification of the capacitance maxima according to the angular momenta is further confirmed by their magnetic-field dependence. The s-shell is only little affected by the magnetic field whereas the p-shell exhibits a magnetic-field dependent splitting, with two peaks decreasing with magnetic field and two increasing for $B < 15\,\mathrm{T}$. For $B > 7\,\mathrm{T}$ two more peaks can be identified showing a negative magnetic-field dependence for $B < 15\,\mathrm{T}$. We associate them to charging of the d-shell with the first two electrons. At around 15 T we observe a magnetic-field-induced intermixing of the p- and d-shell. The magnetic-field dependence of the spectra can be described well by both the Fock model and by a recent many-particle theory by Wojs and Hawrylak. [12,13].

All this information obtained on a sample with a relatively large gate area gives us a complete picture of the evolution of the ground states of self-assembled quantum dots with electron occupation and magnetic field. We can quantitatively explain the dominant features in the measured capacitance spectra by the shell structure of the electron states in these artificial atoms assuming the dots to be equal and non-interacting. A more complex model of the entire dot ensemble and capacitance device, however, is needed to explain an additional fine structure which is observed in the capacitance spectra.

Fig. 2 shows excerpts of capacitance spectra recorded on the same sample, but for different thermal cycles. Only the gate voltage range of s-shell charging is shown for $B = 0$. The trace on top is the same as shown in Fig. 1. It can be seen that the double structure of the s-shell is accompanied by several smaller peaks (indicated by the dotted lines). We observe that thermal cycling only changes the amplitude of this fine structure, the energetic spacing, however, is highly reproducible. The typical distance between these additional maxima is about 30–40 mV in gate voltage, corresponding to roughly 5 meV in energy (lever arm = 7).

For smaller gate areas and therefore smaller numbers of dots, the additional structure becomes more dominant. Fig. 3 shows the CV traces of a sample with a gate area $A = 16\,\mu\mathrm{m}^2$ at different magnetic fields applied in growth direction. The number of dots under the gate is about 1300. The

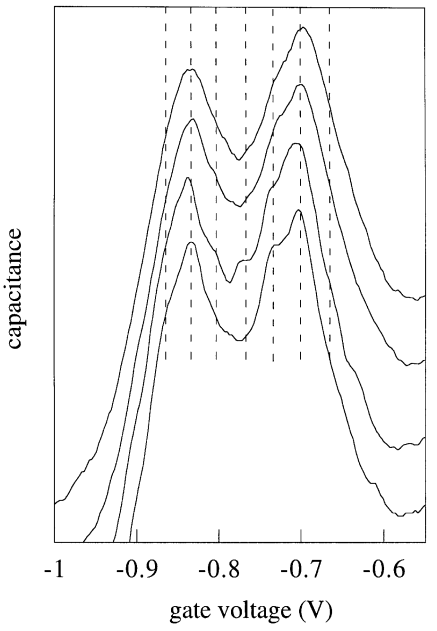

Fig. 2. Differential capacitance for the sample with $A = 89\,\mu\mathrm{m}^2$ for different thermal cycles. Only the gate voltage range of the s-shell is shown.

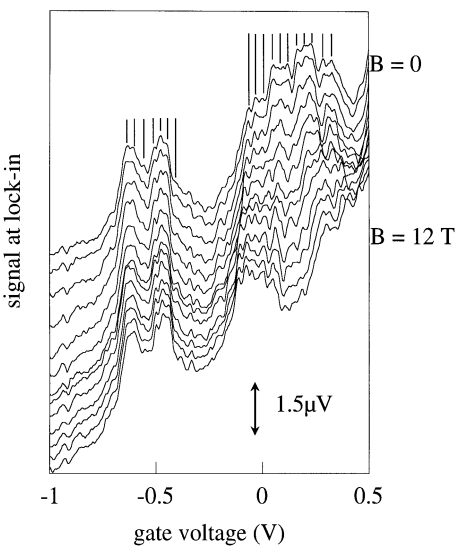

Fig. 3. Differential capacitance for a sample with $A = 16\,\mu\mathrm{m}^2$ at different magnetic fields applied in the growth direction (conversion factor roughly 100 aF/μV). From top to bottom the field is increased in steps of 1 T from $B = 0$ to 12 T.

gray-scale plot in Fig. 4 depicts the capacitance as a function of gate voltage and magnetic field. It can be seen from both Figs. 3 and 4 that the additional maxima within the s-, p- and d-shell basically show the same magnetic-field dependence as the main charging peaks. The energetic spacing between the additional peaks is again roughly 5 meV. The typical broadening is only about 5 meV (as compared to the 15 meV broadening of the s-shell charging peak).

An investigation of the correlation between these additional peaks reveals that they can be grouped into sets. Each of these sets is essentially a replica of the spectrum of the main maxima shifted by roughly 5 meV or multiples thereof. All of these effects imply that the device capacitance contains replicas of the quantum dot density of states, energetically shifted with respect to each other.

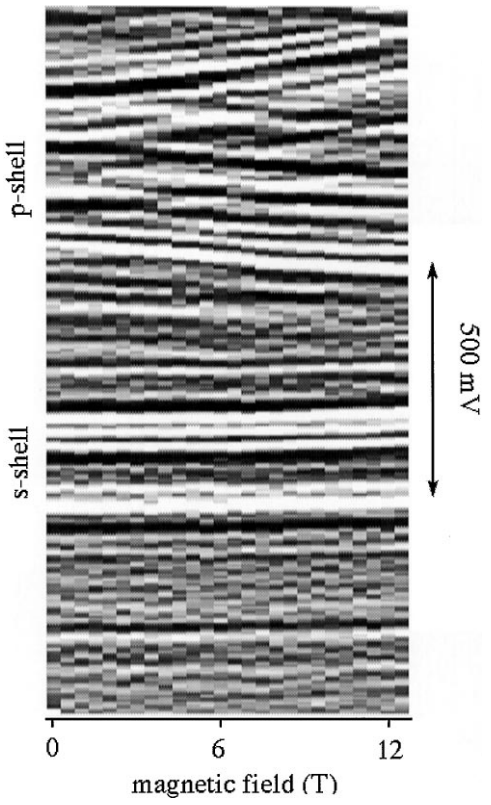

Fig. 4. Gray-scale plot of the same sample as in Fig. 3. White areas correspond to maxima, black areas to minima in the capacitance.

It is difficult at this stage to say with any certainty whether the fine structure is a general property of self-assembled quantum dots or whether the dots are sampling fluctuations in the capacitance device itself, caused, e.g. by neighboring charges or local fluctuations of the lever arm.

One likely explanation in terms of dot property is monolayer fluctuations in the dot heights. Numerical calculations of the ground state energies of our dots with different heights show that a change of one monolayer in height corresponds to an offset of 3–4 meV in energy [14]. The lateral quantization energy will only change by about 1% per monolayer, if we assume disk-shaped dots. Therefore, the magnetic-field dependence of energy levels for dots with different heights should be essentially identical with the threshold gate voltage offset by about 3–4 meV. The dependence on the cool down, however, is not satisfactorily explained.

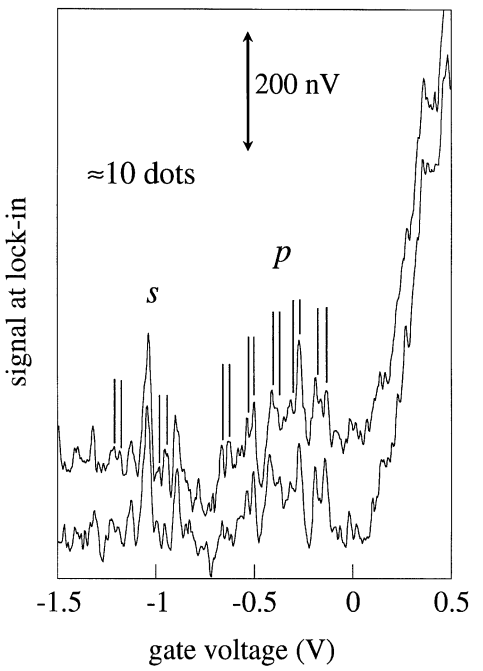

Fig. 5. Differential capacitance for a sample with $A < 0.5\,\mu m^2$ (conversion factor roughtly 24 aF/μV). Very sharp maxima can be observed. Some of them are separated by roughly 5 meV (indicated by lines). Two identical measurements are shown to illustrate the reproducibility of the peaks.

Another possible explanation may be inter-dot coupling. Evaluation of the next-neighbor distance yields a value of about 40–60 nm [15–17]. We estimate the dot–dot interaction at this length scale to about a few meV. To explain the shifted energy spectra one has to include a long-range disorder potential creating domains in the dot ensemble. Charging of the dots within these domains will be influenced and ordered by this Coulomb term.

For a better understanding of the amplitude of the fine structure with respect to the main charging peaks and background, we have attempted to study even smaller dot ensembles. Recent measurements on a sample with a gate area $A < 0.5\,\mu m^2$ show sharp reproducible peaks in the capacitance spectra (see Fig. 5). About 10–20 dots are investigated in this sample. According to their magnetic-field dependence the peaks are named s- and p-like again. They are typically broadened by 4–5 meV (the modulation amplitude accounts to 1–2 meV) with a height of about a few aF. From the integral of the CV characteristic we estimate the number of electrons per maximum to be about 2–4. Some of the maxima are separated again by roughly 5 meV (indicated by lines). It seems to indicate that in the limit of very few dots fine structure-like features dominate the capacitance spectra.

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