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Many-particle ground states and excitations in nanometer-size quantum structures

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Abstract

Recent experiments, which study the electronic structure of self-organized InAs nanostructures, are summarized. The results from simultaneously performed far-infrared (FIR) and capacitance spectroscopy allow for a detailed investigation of both many-particle ground states and excitations in these three-dimensionally confined, few-electron systems. Experimental data from quantum dots, coupled dots and ring-like structures are presented and evaluated with respect to the contributions from quantization, electron–electron interaction and an external applied magnetic field to the energy spectrum. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: Self-organized nanostructures; Quantum dots; Quantum rings; Spectroscopy

1. Introduction

The so-called Stranski–Krastanow (S-K) growth mode has been known to exist in strained semiconductor heteroepitaxy for decades [1]. However, only recently it has been realized that self-organized S-K-islands provide for almost ideal systems to study the electrical and optical properties of dislocation-free, nm-size semiconductor quantum dots [2–4]. These systems are of great interest, not only for studying the basic properties of man-made 'artificial atoms', but also because of possible applications, ranging from novel computing schemes [5] to thresholdless solid state lasers [6]. A deep knowledge of the internal electronic structure in self-assembled quantum dots is therefore highly desirable.

Here, we study the many-particle electronic states and excitations of InAs quantum dots, embedded into a GaAs/AlGaAs heterostructure which allows for a controlled tuning of the quantum dot energy with respect to a reservoir. Using capacitance and far-infrared spectroscopy, we can simultaneously study both the many-particle ground states as well as the excitations of the dots. This way, we obtain an in-depth picture of the contributions of confinement, electron–electron interaction and the influence of an external applied field to the energy structure of these few-electron systems. Making use of improved growth technologies, we can apply the same spectroscopic techniques to investigate the electronic structure of more complex nanostructures, such as double dots or ring-shaped islands.

2. Experimental – dots

The samples are grown by solid-source molecular beam epitaxy. Details of the growth procedure can be found, e.g., in Refs. [2,7]. Inside

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a field-effect-transistor structure between a highly doped GaAs layer, serving as a back contact, and a surface Schottky gate, the InAs dots are embedded in nominally undoped GaAs (see Fig. 1). Approximately 1.7 monolayers of InAs are deposited at 530°C which results in islands of ≈20 nm diameter and ≈6 nm height, with a density of around 10^{10} cm^{-2}. The distance between the dots and the back contact is small enough to allow for tunneling, so that in general, the dots are in equilibrium with the back contact. Because of their large distance from the Schottky gate and an inserted AlGaAs blocking barrier, no charge transfer between the dots and the top gate is possible in the range of voltages investigated. A schematic of the resulting conduction band structure is sketched in Fig. 1(b). The electric field between the back contact and the gate is given by the Schottky barrier and the applied gate voltage V_g. By changing V_g, the energy of the dot with respect to the back contact can be adjusted. Assuming that the charge in the dot layer is negligible, the energy shift is directly proportional to the gate voltage [8]. The proportionality factor is given by the 'lever arm', i.e. the ratio between the tunneling barrier thickness t_{tun} and total thickness t_{tot}, $\Delta E = eV_g \cdot t_{tun}/t_{tot}$ (cf. Fig. 1(b)). For appropriately chosen sample dimensions, the energy of the dots can be shifted so that for moderately negative voltages (≈−1 V), the dots are void of electrons and with increasing bias can be charged with upto 8 electrons. The exact voltages at which additional electrons can tunnel into the dots can be monitored by superimposing upon V_g a small modulation voltage V_{mod}, which will cause the electron to oscillate between the back contact and the dot. This alternating current is detected in the outside circuit as an additional capacitive signal.

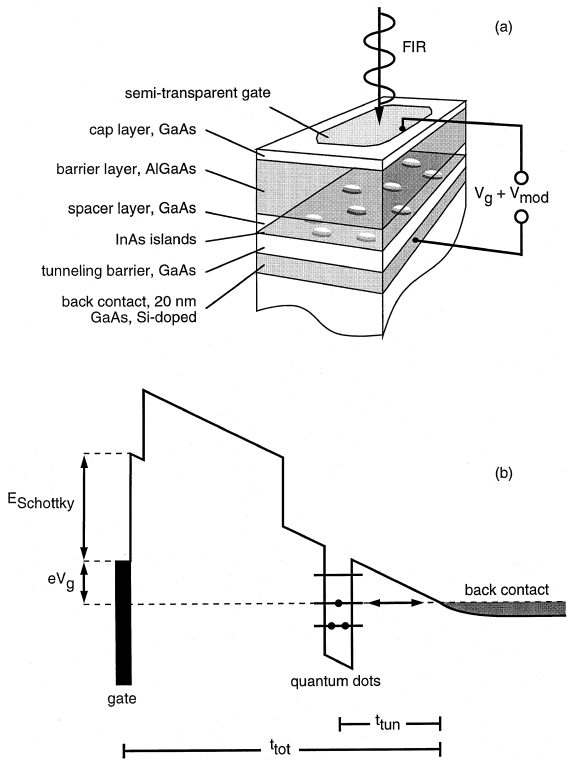

Fig. 1. (a) Schematic of the measurement geometry and layer sequence of the field-effect transistor structure with the embedded InAs quantum dots. (b) Sketch of the conduction band edge of the device. For details, see text.

Fig. 2 shows capacitance–voltage (CV) traces of a sample with an area of ≈0.2 mm^2 and a lever arm of 1/7. From bottom to top the magnetic field is raised from $B = 0$ to 12 T in steps of 2 T. Around $V_g = -1.1$ V two distinct maxima can be observed with a position which is almost independent of magnetic field. Between $V_g = -0.6$ V and

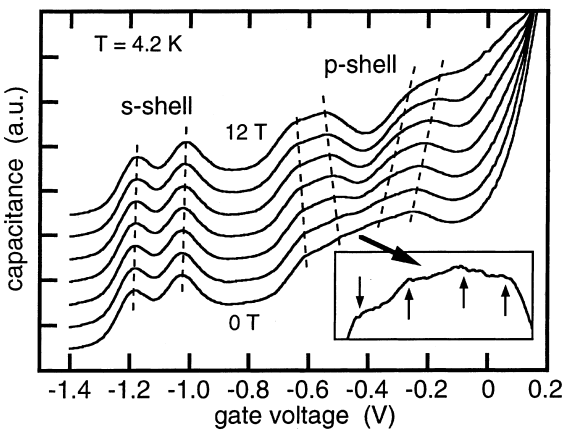

Fig. 2. Charging spectra of self-organized InAs quantum dots. Individual charging peaks are resolved for both s- and p-shells (see also inset). From bottom to top curve, the magnetic field is increased from 0 to 12 T in steps of 2 T, and the orbital Zeeman splitting of the p-shell is clearly observed. Curves are offset for clarity.

$V_g = -0.1$ V a broad shoulder appears which for the present, very homogeneous sample, is seen to consist of four, roughly equidistant peaks. These observations are consistent with a simple picture of the many-particle states in the investigated quantum dots: For their oblate, nearly circular geometry, the lowest and second lowest single-particle states are expected to be doubly and fourfold degenerate, respectively. For the many-particle states, this degeneracy is lifted by electron–electron interactions, so that groups of charging peaks ('shells') appear [9]. This picture agrees with the observed CV spectrum shown in Fig. 2. In analogy with atomic physics, the two lowest states are commonly labelled 's-shell', the next four higher states 'p-shell'. When a magnetic field is applied perpendicularly to the plane of the dots, the angular momentum degeneracy of the p-shell is lifted and at sufficiently high fields the two states with angular momentum $l = +1$ shift to higher energies, whereas the states with $l = -1$ decrease in energy (see Fig. 2). At low fields, a close examination of the p-states reveals the influence of the exchange interaction [10,11]: According to Hund's rules, at $B = 0$ the first two levels of the p-shell which become occupied have different spatial quantum numbers. The angular momentum of the second p-state changes from $l = +1$ to $l = -1$ when the magnetic energy becomes dominant over the exchange contribution (around 1 T for the present samples).

Fig. 3 displays the normalized far-infrared (FIR) transmission through a similar sample at a gate voltage where approximately two electrons occupy each dot. At $B = 0$, a single [12] resonance is observed which splits into two as a magnetic field is applied. This splitting is the dynamic analogue of the orbital Zeeman spitting observed in Fig. 2. In a single-particle picture it can be understood as the two possible transitions between the $l = 0$ s-states and the $l = \pm 1$ p-states, excited by the left- and right-circularly polarized components of the FIR radiation.

The magnetic-field dispersion of the two modes, commonly labelled 'ω_+-' and 'ω_--mode' can very well be described using a parabolic confining potential [13]. It is a peculiarity of the parabolic potential that the excitation energies are independent of electron–electron interactions [14]. Indeed,

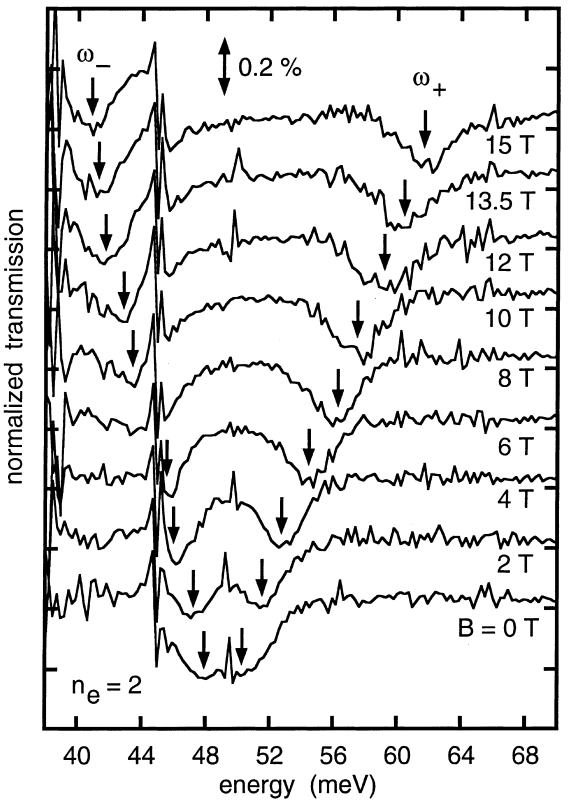

Fig. 3. FIR spectra of self-organized dots (occupied with $n_e \approx 2$ electrons each) for different magnetic fields. The well-known, two-mode spectrum of parabolically confined quantum dots is observed. Curves are offset for clarity.

in many man-made quantum-dots, this leads to the fact that the observed resonance positions are independent of electron number [15,16], ranging from the single electron limit to the quasi-classical situation where dozens or even hundreds of electrons occupy each dot. Also for the present dots, no significant change in energy can be observed when the number of electrons per dot is doubled from $n_e = 1$ to $n_e = 2$. This further supports the assumption that, at least for low occupation, the dots' confining potential is to good approximation parabolic.

This situation changes drastically, however, when n_e is further increased and the p-shell becomes occupied. As seen in Fig. 4, up to three ω_+-like modes appear for a partly occupied p-shell. Interestingly, for even higher occupation,

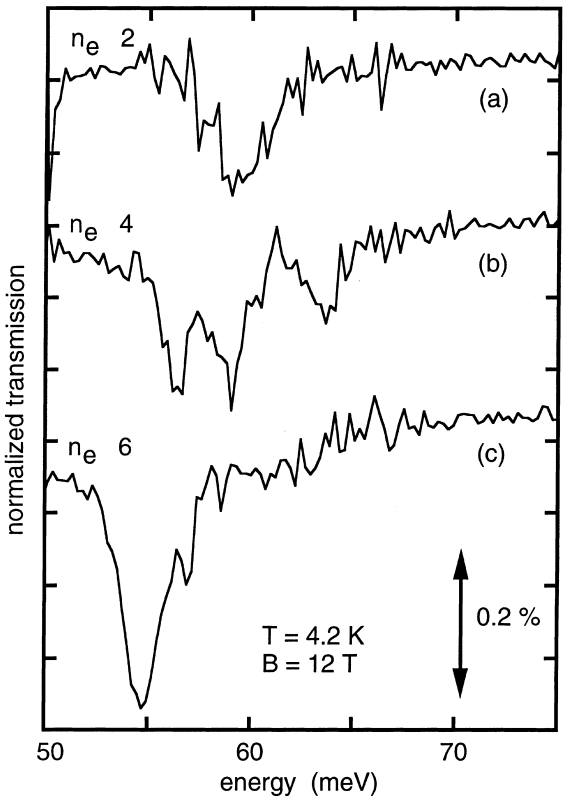

Fig. 4. FIR excitation spectra at $B = 12$ T for different occupation numbers n_e. For a partly occupied p-shell ($n_e = 4$), a splitting of the ω_+-mode into three resonances is observed.

when the p-shell becomes fully occupied and the d-shell starts filling up, the spectrum becomes less featured and only a single ω_+-resonance remains (Fig. 4(c)), however with a dispersion that differs strongly from the parabolic model [3,13]. This way, the strong non-parabolic contributions to the confining potential of self-organized InAs quantum dots make it possible to perform a spectrosopy of the quantum dot 'table of elements' and distinguish, e.g. 'quantum dot helium' and 'quantum dot beryllium' by their respective spectroscopic fingerprints.

3. Coupled dots

In the above experiments, the dots are fairly dilute, so that they can to good approximation be treated as non-interacting. For the investigation of *coupled* dots, a further self-organization mechanism, found in S-K growth, is beneficial. Studies of multi-dot-layers have shown that strain-effects can lead to a vertical alignment of the dots in the different layers [1,17]. It is thus possible to grow two narrowly spaced layers of dots where the vertical alignment leads to molecule-like coupled double dots. So far, we have only investigated double dots with relatively large spacers (10–20 nm), because no well-defined structures with thinner layers could be fabricated (the reason for this might be the morphological changes that can occur when the dots are covered with thin layers of GaAs, see below). Therefore, the main coupling mechanism between the dots is Coulomb interaction rather than quantum mechanical lifting of level degeneracy [18]. There are, however, first indications of quantum coupling effects in our samples, which reveal themselves in the charging *dynamics*: When the levels in both dot layers are aligned, tunneling into the second dot layer from the back contact is orders of magnitude faster than when they are out of resonance [19].

Fig. 5 shows a CV spectrum of a double-dot sample with spacing $d = 20$ nm (thick solid line). The lever arm for the first dot layer is 1/7, identical to the lever arm of a *single*-dot reference sample,

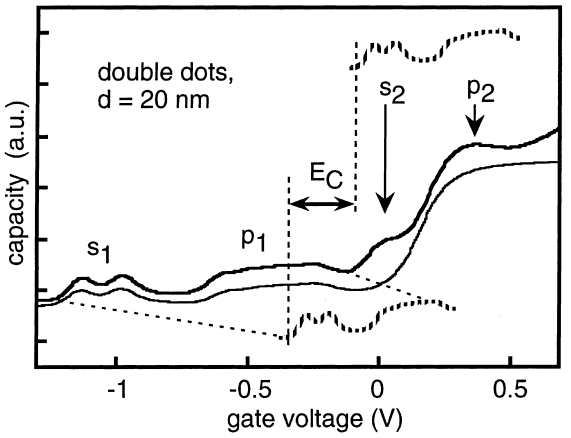

Fig. 5. CV spectrum of a double-dot-layer sample (thick solid line) compared to that of a single-layer structure (thin solid line). In the double-layer sample, two additional structures are observed (arrows), which are attributed to the charging of the second dot layer.

whose CV trace is shown as a thin solid line for comparison. Up to $V_g = -0.2$ V, the two traces are almost identical, demonstrating the nice reproducibility of the growth procedure. Around $V_g = +0.1$ V, when in the single-dot sample the gap between the p-states and the loading InAs wetting layer [20] is observed, a clear peak appears in the double-dot system. An additional, broader structure is resolved around +0.3 V.

From our studies of single dots, we know that the lever-arm conversion between gate voltage and energy holds to good approximation for the present, low-dot-density samples. Since the lever arm of the second dot layer is only 1/3.9, we expect the spectrum of the second dot layer to be shifted and compressed with respect to that of the first layer. To illustrate this, an appropriately scaled and shifted spectrum is displayed at the bottom of the figure (thick dotted line). The clear shift between the scaled spectrum and the observed maxima from the second dot layer is attributed to an additional Coulomb energy contribution E_C, as indicated in the figure. An in-depth, three-dimensional treatment of the interaction between the dot systems is not available at present. However, a first evaluation of the interaction energy, using a model of point charges, can reproduce the measured E_C with satisfactory accuracy [21].

By varying the spacer thickness and the number of stacked dot layers, a large number of novel structures, ranging from weakly coupled dots to quasi-one-dimensional chains of strongly interacting islands can be realized. Furthermore, the fact that with increasing number of stacked layers the *lateral* ordering increases [17], opens new routes to creating two- or three-dimensional, ordered arrays of coupled quantum structures.

4. Ring-like structures

Additional flexibility in the fabrication of self-organized InAs islands comes from a controllable change in island shape after the InAs has been covered with GaAs host material [22]. An example of such a morphological change is shown in Fig. 6 (inset). The atomic force micrograph displays InAs islands which have been covered with 4 nm of GaAs, followed by an annealing step at the growth temperature of 530°C. Strikingly, the shape of the dots has completely changed and is now ring-like. The inner diameter of the rings is approximately 30 nm, the outer diameter ≈80 nm, with a slight elongation along the [1,−1,0] direction. It should be pointed out that this change in morphology is a result of the growth interruption only. On one hand this is interesting from the materials point of view, since it indicates that, at least for covered dots, the shape of common dots (grown without growth interruption) is not given by equilibrium properties. On the other hand, it makes it possible to grow samples with identical nominal layer sequence, with only the shape of the InAs islands altered by the growth interruption.

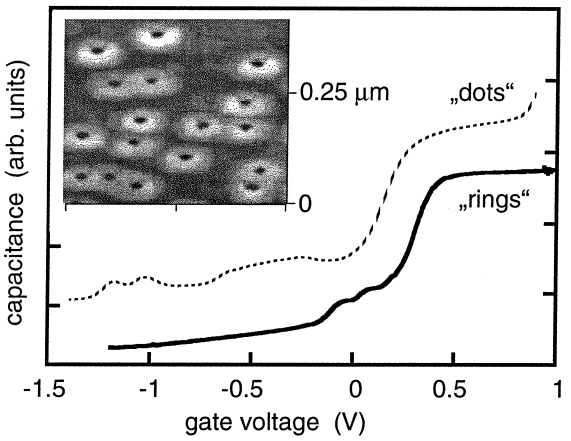

Fig. 6. Charging characteristic of self-organized 'rings', compared to the CV spectrum of conventional dots. The inset displays an atomic force micrograph of the uncovered ring structures.

Fig. 6 shows that also the electronic properties inside the final structure are strongly affected by the growth interruption. The solid line displays the CV spectrum of a 'ring'-sample, compared to that of a 'dot'-sample with identical layer sequence (dotted line). Instead of the typical s- and p-shell characteristic of the 'dot'-structure, only two maxima can be observed for the 'ring'-sample. Their absolute and relative strengths as well as their negligible magnetic-field dependence lead us to conclude that they represent the filling of the first and the second electron level. Their spacing corresponds to 19 meV, almost identical to the s-split-

ting of the 'dot'-sample (22 meV). The far-infrared response of the 'rings', on the other hand, is drastically different from that of the dots (Fig. 7). Even though a two-mode behavior, similar to that of quantum dots, can be observed (solid lines), the $B=0$ resonance is reduced to 20 meV, less than 50% of the excitation energy of the dots. Even more striking is the appearance of two additional modes, ω_1 and ω_2, which extrapolate to very low frequencies for vanishing magnetic field. Phenomenologically, their magnetic-field dispersion can be described by $\omega_1=\omega_c/2+\Delta_1$ and $\omega_2=\omega_c+\Delta_2$ (dotted lines in Fig. 7), with $\Delta_1\approx\Delta_2\approx 7$ meV/$\hbar$. Here, $\omega_c=eB/m^*$ is the cyclotron frequency, taken from the Zeeman splitting between the ω_+- and ω_--modes of both the 'dots' and the 'rings'.

Although the morphology of the rings shown is Fig. 6 is unambiguous and has been confirmed by atomic force and scanning electron microscopy as well as small-angle X-ray scattering [23], care should be taken when assigning the observed resonances to a true ring geometry. The annealed islands shown in Fig. 6 have to be further covered in order to complete the necessary layer structure (cf. Fig. 1(a)), and the additional deposited material might result in a further shape change. A thorough evaluation of the CV- and FIR-spectra is therefore of great value in determining how far the electronic structure inside the completed samples is indeed ring-like. Such an evaluation is presently underway.

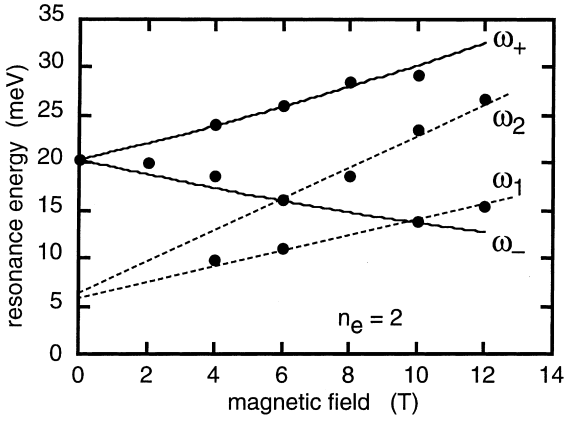

Fig. 7. FIR resonance positions of self-organized 'rings' as a function of magnetic field for occupation number $n_e=2$. In addition to the two-mode spectrum (solid lines), two novel resonances appear.

First conclusions, however, can be drawn in view of the experimental data. In general, Coulomb contributions roughly scale as $1/L$ (L being a characteristic length of the system), whereas quantization scales as $1/L^2$. Since the latter is expected to dominate the excitation spectrum, the observation of a much lower $B=0$ resonance is in agreement with the increased size of the 'rings' with respect to the dots. Applying the above scaling argument, a Coulomb interaction energy of only 13 meV is expected, much lower than the observed value. Thus the electronic properties suggest that not only a change in *size* but also in *shape* of the InAs islands is induced by the annealing step, and that this morphological change is preserved during further overgrowth.

In conclusion, the S-K growth procedure allows for the fabrication of a number different nm-size structures, whose intricate electronic structure can be investigated in great detail, using combined FIR and CV spectroscopy. Even though the self-organized nature of this fabrication technique seems to imply a limited control over the morphology of the resulting nanostructures, a number of interesting quantum systems can be realized, ranging from single and coupled dots to quantum rings. These allow for experimental tests of a number of fundamental concepts and predictions concerning coupling-, quantum-, and size-effects in semiconductor nanostructures.

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