Spectroscopy of Quantum Levels in Charge-Tunable InGaAs Quantum Dots

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Imbedding self-assembled lens-shaped InGaAs quantum dots in a suitably designed field-effect-type GaAs/AlAs heterostructure allows us to charge the lowest discrete quantum levels in the dots with single electrons. Because of their small diameters of about 20 nm the Coulomb charging energy is significantly smaller than the quantization energies. We extract energy spacings of about 41 meV between the *s*-like ground state and the first excited *p*-like state from capacitance as well as infrared transmission spectroscopy at low temperatures and under application of high magnetic fields.

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Quantum dots in semiconductors are fascinating man-made objects with potential device applications both as electronic memories as well as optoelectronic devices. Various schemes have been developed to create confining potentials in which electrons and possibly holes are quantum confined in all three spatial directions. The most extensively studied quantum dots use electrostatic confinement in GaAs employing suitably patterned surface topology and gate electrodes [1,2]. In such devices typical lateral confinement lengths are 100 nm or larger, and correspondingly Coulomb charging energies often dominate quantization energies when single electrons are added to the quantum dots. Furthermore, the bare lateral potentials are nearly parabolic since rather remote surface and gate charges define the confining potential. This makes infrared (IR) spectroscopy of the confined electrons insensitive to the discrete nature of the quantum-confined states as a consequence of the so-called generalized Kohn theorem [2]. Alternative schemes that try to completely confine electrons and holes by suitable growth and patterning techniques have been able to produce complex and intriguing luminescence spectra [3–5] but have not yet resulted in quantum dots in which the occupation of individual quantum levels with electrons can be easily controlled.

Here we report on the realization of such atomlike dots which combine confinement of both electrons and holes with tunability of the electronic charge as well as large confinement energies desired for applications. Imbedding InGaAs quantum dots generated by coherent island growth [6,7] in a suitably designed MISFET (metal-insulator-semiconductor-field-effect-transistor)-type GaAs/AlAs heterostructure allows us to controllably inject single electrons into the quantum dots and to occupy the lowest quantum confined states. With capacitance spectroscopy [8,9] we study the discrete density of states (DOS) of the quantum dot levels. With IR spectroscopy we directly observe dipole-allowed transitions between levels of the confined electrons with energies that compare quantitatively with those extracted from the capacitance spectra. Our devices thus realize charge-tunable artificial atoms and are excellently suited to study effects of electron occupation and electron-electron interactions.

The heterostructures studied here are grown by molecular beam epitaxy with the essential layer sequence as sketched in Fig. 1. On a semi-insulating GaAs (100) substrate with a suitable buffer a 20 nm thick n^+-doped (Si, 4×10^{18} cm^{-3}) GaAs layer is grown serving as a back contact. On top of an undoped GaAs layer (thickness z_b = 50 nm) the InGaAs quantum dots are formed by growing a coherently strained $In_{0.5}Ga_{0.5}As$ layer. The growth is interrupted at the transition from two-dimensional (2D) to three-dimensional growth as judged by the reflection high-energy electron diffraction (RHEED) pattern [6]. At this point dislocation free, lens-shaped InGaAs dots, so-called Stranski-Krastanow islands [10] are spontaneously formed. The dots are statistically distributed within the interface plane with their density (typically 10^{10} cm^{-2}) and diameter (typically 20 nm) somewhat adjustable by the choice of

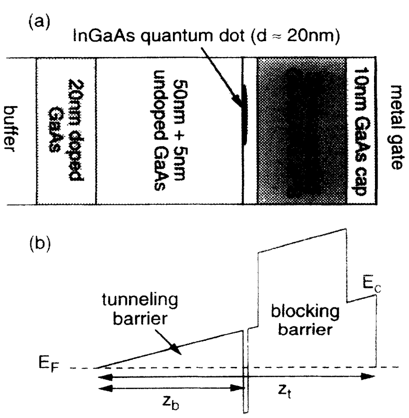

FIG. 1. (a) Layer sequence of our device. The InGaAs dots are statistically distributed within the plane sandwiched by two GaAs layers and have a lateral diameter of about d = 20 nm. (b) Sketch of the conduction band edge E_C with respect to the Fermi level E_F along the growth direction for gate voltages at which no electrons are in the InGaAs dots.

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growth conditions. They are remarkably uniform in size with their diameter fluctuating by only about 10% and their thickness of a few monolayers by only about a single monolayer [6,7]. The dot growth is followed by a 5 nm thick GaAs layer, a 30 nm thick short period AlAs/GaAs superlattice (period 4 nm) serving as blocking barrier and a 10 nm GaAs cap layer. The MISFET-type devices are completed by alloying In contacts to the back contact layer and evaporating a semitransparent NiCr gate electrode. A sketch of the conduction band edge along growth direction at zero gate bias is shown in Fig. 1(b).

With forward bias, electrons can be injected from the back contact into the InGaAs dots through the GaAs tunneling barrier. The capacitance measured at temperature $T = 4.2$ K is displayed in Fig. 2(a) and illustrates the charging characteristics of the device in the presence of a magnetic field B applied perpendicularly to the sample surface. At $B = 0$ T and gate bias below $V_g = 0.32$ V the capacitance reflects the geometric capacitance between the back contact and the gate is determined by the distance z_t. At $V_g = 32$ V the capacitance increases, and electrons are injected into the originally unoccupied quantum dots. As discussed below the two maxima in the capacitance trace reflect the occupation of the lowest two quantum states of the dots. Above about $V_g = 0.6$ V electrons are transferred into the GaAs layer above the dots and accumulate at the interface between the GaAs and the AlAs/GaAs blocking barrier forming a 2D electron gas (2DEG). This can be immediately judged from Shubnikov–de Haas–type quantum oscillations in the magnetocapacitance at voltage above $V_g = 0.6$ V not shown here.

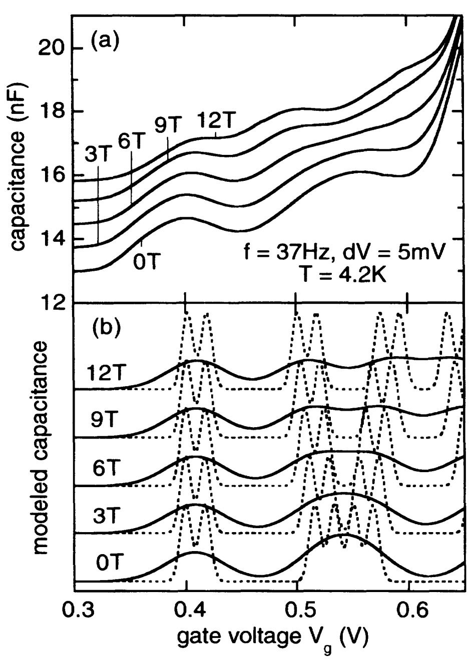

FIG. 2. (a) Measured capacitance as a function of gate voltage V_g for different magnetic fields B increasing in steps of 3 T from the bottom to the top trace. The vertical scale refers to the bottom trace ($B = 0$ T), and a constant offset is added to the other traces for clarity. The frequency of the excitation voltage $dV = 5$ mV is $f = 37$ Hz. (b) Calculated peak positions of the capacitance traces according to Eq. (1). The voltage V_{tz} is chosen to $V_{tz} = 0.31$ V, and the capacitance $C = 9.2$ aF is equal to the self-capacitance of a disk with 20 nm diameter in GaAs ($\varepsilon = 13$). The magnetic field dependence of the energy levels is calculated for a parabolic confining potential [12] with $\hbar\omega_0 = 41$ meV and $m^* = 0.07m_0$ as obtained from the IR transmission experiments shown in Fig. 4. A Gaussian function is assumed for the peaks with a standard deviation of 5 and 25 mV for the dashed and full lines, respectively.

We now want to focus on the two maxima in the magnetocapacitance at intermediate gate voltages. In this regime we can convert the gate voltages at which we observe maxima in the capacitance and hence in the DOS into an energy scale by using the following simple relation:

$$V_g - V_{tz} = (N - 1/2)e/C + (z_t/z_b)E_n/e\,. \qquad (1)$$

Here we assume the lateral quantization energy E_n in the quantum dot to be separable from the much larger quantization energy in growth direction E_z which equals the Fermi level E_F at threshold voltage V_{tz}. The first term on the right-hand side is the Coulomb charging energy required to charge N electrons into the dot capacitance C, and the second term reflects the lateral quantization energy E_n adjusted by the appropriate potential lever arm $z_t/z_b \approx 2$. To estimate the charging energy e^2/C we calculate the self-capacitance $C = 4\varepsilon\varepsilon_0 d$ of a disk-shaped dot of $d = 20$ nm diameter imbedded within bulk GaAs, yielding a charging energy of about 18 meV. Screening by the gate and the back contact will reduce this value somewhat such that the above value can be considered an upper bound. Assuming a Gaussian broadening of the thermodynamic DOS of the quantum dot levels we can simulate the capacitance trace caused by size-induced fluctuation of the dot size if we know the energies and the degeneracies of the relevant quantum levels as illustrated in Fig. 2(b) for two different broadenings. Whereas small broadening would make a resolution of the Coulomb charging of individual electrons visible, a broadening of about 25 mV that results in simulated capacitance spectra quite similar to the measured ones will smear the Coulomb charging maxima but still allows us to well resolve the DOS maxima caused by size quantization.

Assuming spin degeneracy, we thus need about $2E_0/e + 3e/2C$ to fully charge the ground state of the quantum dot. Interpreting the next maximum of the capacitance as reflecting the charging of the fourfold degenerate p-like E_1 state of the quantum dot, we expect a voltage difference of $\Delta V_g = 3.18$ mV $+ 2(E_1 - E_0)/e = 145$ mV between the two peaks. From the IR spectra discussed below we obtain a value of $E_1 - E_0 = 41$ meV, yielding $\Delta V_g = 136$ mV in close correspondence to the value found in capacitance. Comparable energy differences ($E_1 - E_0$) are also deduced from photoluminescence studies of similar InGaAs quantum dots [11] and are consistent with simple models of quantum dot energies. For example, for a spherical quantum disk of 20 nm diameter we calculate surprisingly close values.

We now turn to the analysis of the magnetic field dependence of the two capacitance maxima of interest here. The lowest capacitance maximum shifts only slightly with increasing magnetic fields to higher gate voltages. The upper maximum, however, splits into two distinct peaks, one decreasing in gate voltage and one increasing in gate voltage with increasing magnetic field. Since the confinement is much stronger in the growth direction, we model the lowest quantum states to result from lateral quantization only. For simplicity we assume a parabolic lateral confinement potential which yields an energy level structure at $B = 0$ T in which the s-like E_0 level has twofold spin degeneracy, whereas the p-like E_1 level has a twofold orbital as well as twofold spin degeneracy [12]. In a perpendicularly applied magnetic field the expected spin splitting is too small to be resolved under present experimental conditions, whereas the orbital degeneracy is lifted, and the energy difference between E_1 and E_0 is expected to depend on magnetic field as

$$(E_1 - E_0)/\hbar = \omega_\pm = \sqrt{\omega_0^2 + (\omega_c/2)^2} \pm \omega_c/2, \quad (2)$$

where $\omega_c = eB/m^*$ is the cyclotron frequency and $\hbar\omega_0 = E_1 - E_0$ at $B = 0$ T. This is the behavior observed here. The magnetic field dependence of the measured splitting is well described by $m^* = 0.07m_0$ close to the conduction electron mass in GaAs. This can be expected since the electrons in the quantum dot will have an essential portion of their wave function extended into the surrounding GaAs.

In studies of the effect of the orientation of the magnetic field we find both the E_0 as well as the E_1 maximum to be sensitive to the perpendicular magnetic field only, again reflecting that confinement along the growth direction is much larger than lateral confinement. We observe similar capacitance traces on several samples from different wafers. A reference sample is grown with the same basic structure but the 3.5 monolayers (ML) $In_{0.5}Ga_{0.5}As$ as compared to the 4.5 ML from which the dot layer is formed. On this sample we only observe the capacitance increase at higher gate voltage resulting from formation of the 2DEG at the blocking barrier.

To further assert that we indeed observe the quantum levels of the InGaAs dots in the capacitance spectra, we have investigated the IR transmission with a Fourier transform spectrometer similar to previous studies of electrostatically confined quantum dots [13,14]. In Fig. 3 we display representative traces of the relative IR transmission at a finite magnetic field and at gate voltage V_g divided by the transmission at a gate voltage below V_{tz}. At $V_g \geq 0.4$ V we observe the appearance of a transmission minimum at an energy of about 45 meV with its strength first increasing with V_g. For electron densities of 10^{10} cm^{-2} corresponding to typical dot densities we expect the integrated area of the IR resonances to be about 3.5×10^{-3} meV. This is in good agreement with the observed transmission minima (0.1%) and resonance linewidths (3 meV). At $V_g = 0.5$ V a new resonance appears at lower energies of about 27 meV again increasing in strength with increasing V_g.

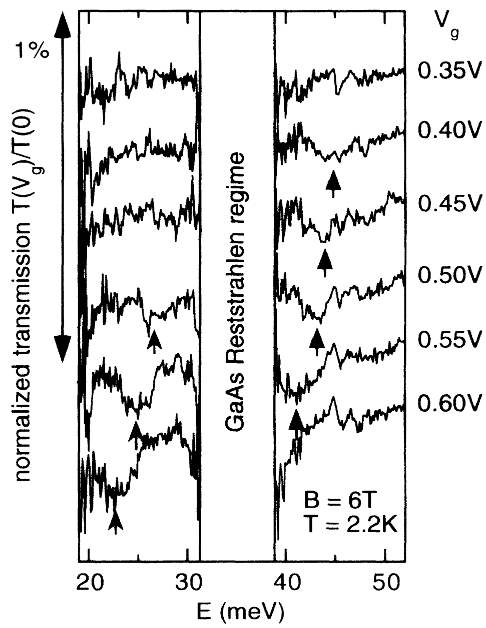

FIG. 3. IR transmission spectra recorded at magnetic field $B = 6$ T and $T = 2.2$ K for different gate voltages V_g. The observed weak resonances are highlighted by arrows.

We find an excellent correlation of the gate voltages at which these weak IR lines appear with the charging peaks in the capacitance. At gate voltages where the E_0 level is seen to be filled in the capacitance, the higher energy IR resonance appears and is thus identified as the $E_0 \rightarrow E_1$ transition. The absorption line at lower energy emerges at gate voltages at which we observe the filling of the lowest E_1 level in the capacitance and hence is interpreted as the $E_1 \rightarrow E_2$ transition. The observation that the $E_1 \rightarrow E_2$ transition is considerably smaller in energy than the $E_0 \rightarrow E_1$ transition can be explained by the fact that the E_2 state is very close to the GaAs conduction band edge and as judged from the capacitance trace possibly even a resonance just above the GaAs conduction band edge. Here, however, a more detailed model of the energy states in the quantum dot that takes the dot shape as well as strains and band offsets into account and possibly includes effects of electron-electron interactions [15] is required to compare quantitatively to the experiment.

The magnetic field dependence of the IR resonances corresponds well to what is expected for electrons in a quantum dot [16]. Figure 4 compares the magnetic field dependence of a parabolic confinement model [Eq. (2)] with the measured resonance position of the $E_0 \rightarrow E_1$ transition. The low frequency ω_- branch predicted by the parabolic model in a magnetic field is not observed here, possibly because it is hidden by the GaAs reststrahlen regime. The magnetic field dependence of the ω_+ branch is well described by a parabolic confining potential with $E_1 - E_0 = 41$ meV and the band edge mass of electrons in GaAs. We note that at voltages above $V_g = 0.6$ V we observe the typical cyclotron resonance of a 2DEG in the IR transmission, confirming that now electrons are forming a 2DEG at the GaAs/AlAs interface in correspondence to the increase in the capacitance signal. For comparison we also have measured the IR transmission through our reference sample with 3.5 ML $In_{0.5}Ga_{0.5}As$. Again we only observe cyclotron resonance of a 2DEG at rather high gate voltages above $V_g = 0.7$ V but no discrete lines at $B = 0$ T and in the gate voltage regime where the dots are charged.

A characteristic feature of the IR dot resonances is that their linewidth of about 3 meV is well below the broadening of the capacitance maxima. This we explain by the fact that size fluctuations of the quantum dot, in particular fluctuations of the dot thickness in growth direction, will predominantly affect the total ground state energy and thus broaden the charging threshold V_{tz} but significantly less influence the energy difference $E_1 - E_0$. Also, we wish to note that similar spectra are obtained on several samples with absolute resonance energies varying only about 10% from sample to sample, even if they are from different wafers. This implies that the dot size is rather stable to inadvertent modifications

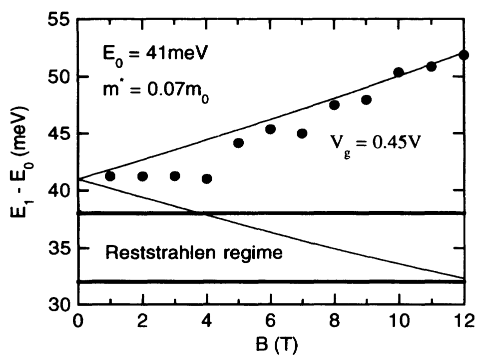

FIG. 4. IR resonance position as a function of magnetic field B at a gate voltage of $V_g = 0.45$ V. The full lines correspond to the transition energies according to Eq. (2) with $\hbar\omega_0 = 41$ meV and $m^* = 0.07m_0$.

in the growth conditions and largely determined by the growth mechanism, in agreement with what is found in recent studies of such quantum dots using an atomic force microscope [17]. This is likely to be the essential reason why we are at all able to observe the discrete nature of the DOS in the quantum dot in capacitance spectra as well as in the IR spectra, though we average in both experiments over a large dot ensemble. This particular property of self-assembling quantum dots may be the essential ingredient needed for their implementation into applicable devices.

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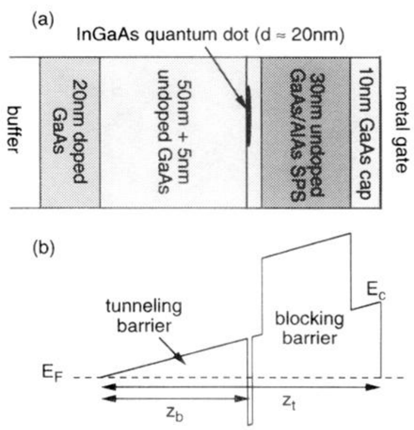

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