

Single-Electron Charging of Quantum-Dot Atoms

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(Received 7 June 1991)

Arrays of field-effect-confined quantum dots with diameters smaller than 100 nm have been prepared starting from $\text{Al}_x\text{Ga}_{1-x}\text{As-GaAs}$ heterostructures. In far-infrared spectroscopy, we induce transitions between the 2-meV-separated quantum levels. We observe discrete steps in the gate-voltage dependence of the integrated absorption strength indicating directly the incremental occupation of each dot with $N=1, 2, 3,$ and 4 electrons. From the gate-voltage dependence, we can estimate a Coulomb charging energy of about 15 meV. On a very fine scale, we also observe a spectral fine structure for the excitation of the quantum-dot atoms.

PACS numbers: 71.45.-d, 73.20.Dx, 73.40.Gk, 73.40.Kp

Low-dimensional quantum confined electronic systems in semiconductors have recently attracted much interest. The ultimate limit is a quantum dot, an artificial atom, where the electrons are confined in all three dimensions [1-6]. We have prepared, starting from two-dimensional electron systems in $\text{Al}_x\text{Ga}_{1-x}\text{As-GaAs}$ heterostructures, high-precision periodic arrays of quantum dots where electrons are confined by the field effect of a laterally structured gate electrode. The separation between the quantized energy levels is typically 2 meV. In far-infrared (FIR) spectroscopy, we excite transitions between these levels and we observe discrete steps in the gate-voltage (V_g) dependence of the integrated absorption strength. Since the integrated absorption strength is proportional to the number of electrons per dot, this indicates directly the stepwise, discrete charging of each dot with 1, 2, 3, and 4 electrons. This links our experiments to another interesting topic, i.e., single-electron charging effects, which are extensively studied in both small metallic and semiconductor systems [7-12]. From the gate-voltage dependence of the steps, we can estimate the Coulomb charging energy to be about 15 meV. This large value stabilizes a well defined number of electrons in each individual dot of the array and allows us to study the spectral fine structure of quantum-dot atoms.

The quantum-dot samples are sketched in the inset of Fig. 1(c). They have been prepared starting from $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As-GaAs}$ heterostructures grown by molecular-beam epitaxy (MBE). The thickness of the spacer layer was 36 nm, that of the doped AlGaAs layer was 56 nm, and that of the cap GaAs layer was 9 nm. A Si δ -doped layer in the GaAs, deposited at a distance of 330 nm from the AlGaAs-GaAs interface, acts as a back contact to charge the dots. The doping density of $2 \times 10^{12} \text{ cm}^{-2}$ was optimized to have enough conductivity for charging the dots but still being semitransparent for FIR radiation. The 2D density was about $N_s = 2 \times 10^{11} \text{ cm}^{-2}$ with a mobility of about $900000 \text{ cm}^2/\text{Vs}$ (at 2.2 K). On top of the heterostructure we prepared a periodic photoresist dot array by holographic lithography. The periods ranged from $a = 500 \text{ nm}$ down to 200 nm and the

lateral photoresist dot sizes were about half the period with a height of about 100 nm. An 8-nm-thick semitransparent NiCr gate of 4 μm diam was evaporated onto the photoresist structure. Contacts were alloyed to the δ -doped back contact, so that with a negative gate voltage we could confine the electrons under the photoresist dots and vary the number of electrons [2-4,6]. FIR transmission spectroscopy was carried out with a Fourier transform spectrometer and with FIR lasers in perpendicular magnetic fields B . We recorded the normalized transmission of unpolarized radiation, $T(V_g)/T(V_t)$, where V_t is the threshold voltage at which the dots are totally depleted.

Experimental FIR transmission spectra for a sample with period $a = 200 \text{ nm}$ are displayed in Figs. 1(a) and 2. In Fig. 1(a) we observe for the laser frequency of 10.5 meV (84 cm^{-1}) resonances at about $B = 6 \text{ T}$. In the frequency-sweep experiment at fixed $B = 3 \text{ T}$ in Fig. 2 resonant absorption occurs at $\omega \approx 44 \text{ cm}^{-1}$. If we follow the FIR resonances at a fixed V_g in their dependence on B , we find a dispersion as shown in Fig. 1(c). This dispersion has been observed previously [4-6]. It consists at $B = 0$ of one resonance, ω_0 [with $\omega_0 = 13 \text{ cm}^{-1}$ (1.6 meV) in Fig. 1(c)], which splits for $B > 0$ into two resonances: One increases in frequency with increasing B and approaches the cyclotron frequency $\omega_c = eB/m^*$ and the other decreases with B (m^* is the effective mass). These B dispersions directly reflect dipole transitions of electrons in a confinement potential of parabolic shape [13],

$$\omega_{r\pm} = \pm \omega_c/2 + [(\omega_c/2)^2 + \omega_0^2]^{1/2}, \quad (1)$$

where ω_0 is the separation of the energy levels in the dot and the excitation frequency at $B = 0$, which is determined by the curvature of the potential. The interesting observation in Fig. 1(a) is that the absorption amplitude does not increase in proportion to $V_g - V_t$. As we will elaborate in the following, this directly indicates that the quantum dots are occupied with a well-defined number of electrons in different V_g intervals. A closer look at the spectra shows that not only the amplitude but also the line shape varies with V_g . To understand this behavior,

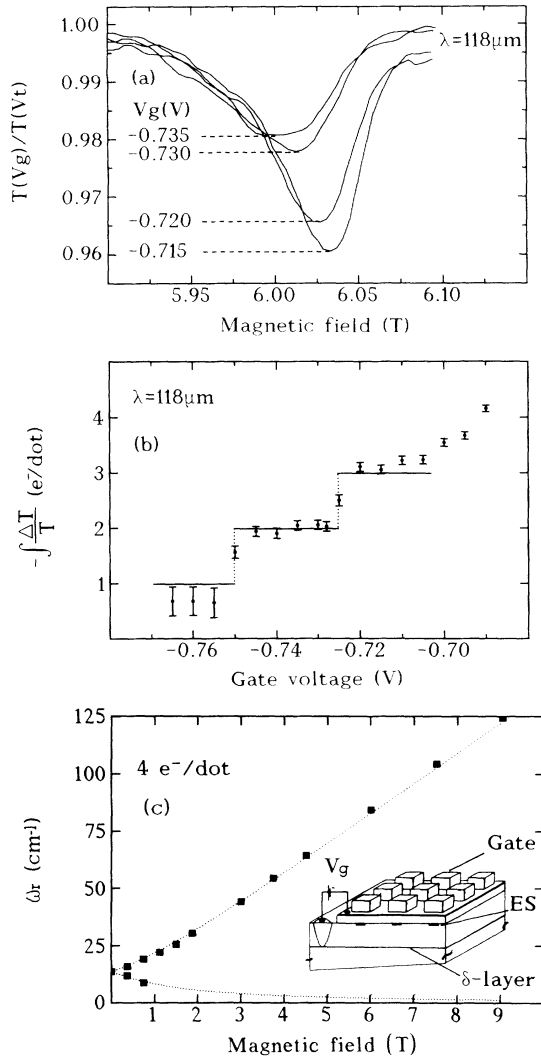


FIG. 1. (a) FIR transmission measured at a fixed laser frequency of 10.5 meV ($\lambda=118 \mu\text{m}$) at various gate voltages V_g for a quantum-dot array with period $a=200 \text{ nm}$. (b) Integrated absorption strength vs gate voltage V_g for a series of spectra including (a)]. The stepwise increase of the absorption strength indicates the incremental occupation of the dots with $N=1, 2,$ and $3 e^-/\text{dot}$ with increasing V_g . (Error bars mark the accuracy of the fits, see text.) (c) B dispersion of the resonance frequencies for a gate voltage where each dot is occupied by $N=4$ electrons. The inset sketches the quantum-dot sample. ES is the position of the electronic system. The temperature is $T=1.2 \text{ K}$.

we have performed a large series of measurements on different samples and in different B regimes. In Fig. 2, we have varied V_g in small increments and we observe that the line shape changes significantly, sometimes consisting of two clearly resolved resonances.

To evaluate these spectra we apply a classical model for the FIR response of electrons in a parabolic potential which have a high-frequency conductivity of [14]

$$\sigma(\omega) = Ne^2\tau / \{2m^*a^2[1 + i(\omega^2 - \omega_r^2)\tau/\omega]\}, \quad (2)$$

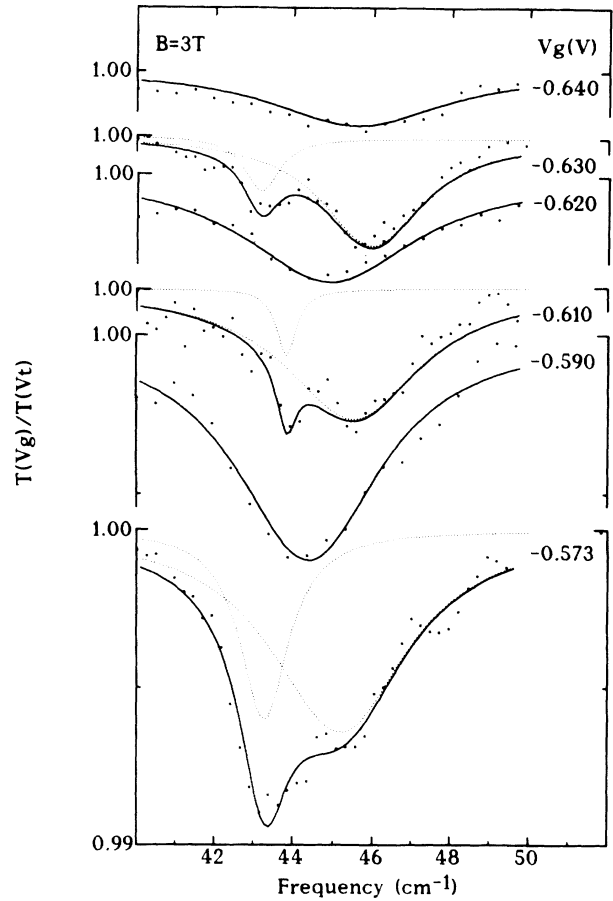


FIG. 2. Experimental FIR transmission spectra (symbols) and fits (solid lines) with two resonances (dotted lines) at $B=3 \text{ T}$ and various gate voltages V_g as indicated. Different spectra are shifted vertically. For some V_g a clear splitting into two resonances is observed. The spectral resolution is 0.25 cm^{-1} and the temperature is $T=2.2 \text{ K}$.

where ω_r is the high-frequency resonance in Eq. (1), N the number of electrons per dot, a the period, and τ a phenomenological scattering time that describes the linewidth. The relative change in transmission $\Delta T/T = [T(V_g) - T(V_t)]/T(V_t)$ is then given by the well-known expression [4,5,14],

$$\Delta T/T = -2 \text{Re}[\sigma(\omega)] / [(1 + \sqrt{\epsilon} + r_c/r_g)\epsilon_0 c], \quad (3)$$

where $\epsilon=12$ is the dielectric function of GaAs, $r_c=377 \Omega$ is the vacuum impedance, and $r_g \approx 1 \text{ k}\Omega$ is the combined impedance of front gate and back contact. Note that the integrated classical absorption strength is identical with the quantum-mechanical result [15]. We can thus determine directly the absolute number of electrons per dot from the experimental signal strength $\Delta T/T$.

We have fitted the spectra in Figs. 1(a) and 2 with the ansatz (2) and (3) using a superposition of two resonances with frequencies ω_{r1} and ω_{r2} . The results are plotted in Figs. 1(b) and 3 and demonstrate that we can directly determine the number of electrons from the FIR

absorption strength. For example, in Fig. 3(a), we find that, above V_t up to -0.64 V, the dots contain $N=1$ electron; between -0.63 and -0.61 V, $N=2$ electrons; between -0.60 and -0.58 V, $N=3$ electrons; and between -0.57 and -0.555 V, $N=4$ electrons. [Note that there is a shift of the threshold voltage in Figs. 2 and 3 with respect to Fig. 1(b). This arises from a different number of electrons transferred from the δ layer during the faster cooldown and the lower temperature, $T=1.2$ K, in the laser-cryostat, resulting in a higher 2D density at $V_g=0$. This, however, does not change the lengths of the V_g intervals.]

It is surprising that for our large number of about 10^8 dots we can charge all the dots simultaneously with the same number of electrons [within the error bars given in Figs. 1(b) and 3(a)]. The reason is the high Coulomb charging energy in these dots which we can estimate from our experiments. From Fig. 1(b) we can see that it requires an increase in the voltage of $\Delta V_g=30$ mV to increment the number of electrons from $N=2$ to $N=3$. This corresponds to a capacitance of $C=e/\Delta V_g=5.3 \times 10^{-18}$ F and a "Coulomb charging energy," defined as $E_c=e^2/2C$, of 15 meV. This value is significantly larger than kT and obviously also larger than local fluctuations of V_t .

Note that our definition of the Coulomb charging energy is necessarily somewhat different from that in classical metallic systems where the capacitance is fixed [7,8]. When we charge the quantum dot with one additional electron, we increase the electronic diameter of the dot and the capacitance. Hence we decrease the Coulomb energy with increasing V_g . It is not easy to calculate directly the capacitance for our complicated geometry.

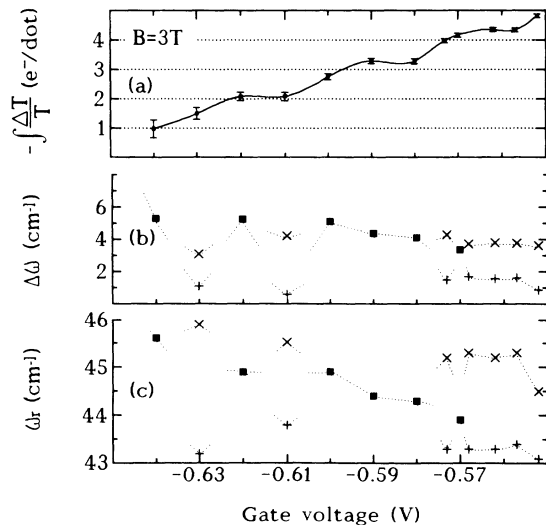


FIG. 3. (a) Number of electrons per dot evaluated from the FIR absorption strength (spectra in Fig. 2). (b) FWHM half-width $\Delta\omega$ and (c) position of the resonance frequencies ω_{r1} and ω_{r2} . The alternating symbols indicate the different V_g regimes where one (■) or two resonances (+, ×) are observed.

However, if we use the model of a parallel-plate capacitor, describing the effective dot area by the extent of the wave function in a harmonic oscillator (54 nm for $N=2$), and take the distance to both the front gate and the back contact layer, we find roughly the same value for C as it is determined from $C=e/\Delta V_g$. Self-consistent Hartree calculation for quantum-dot structures in GaAs with nearly the same dimensions as discussed here have been performed by Kumar, Laux, and Stern [16]. One finds that the voltage intervals required to change the charge of the dot with the equivalent of one additional electron is $\Delta V_g=15$ mV. This value is very similar to ours and confirms our results.

Coulomb charging of quantum dots with few electrons has been discussed, e.g., by Kumar, Laux, and Stern [16], Silsbee and Ashoori [10], and also very recently by Ashoori *et al.* [11]. At this point we would like to demonstrate how the Coulomb charging energy can be estimated in a totally independent "atomic model." This can be done on the basis of many-body calculations of quantum dots, as has been performed, e.g., by Merkt, Huser, and Wagner [17] for two electrons in a parabolic confinement. From their calculations one can deduce that the increase of the two-electron ground-state energy with respect to the one-electron ground state, due to the Coulomb interaction, is $E_c=e^2/\epsilon l_0$. [$l_0=(\hbar/m^* \omega_0)^{1/2}$ is the harmonic oscillator radius.] This gives $E_c=6$ meV for our sample. This energy for the charging from one to two electrons is of the same order as the values estimated from our experimental findings (15 meV for the most clearly resolved 2-to-3 electron charging) and confirms our interpretation. Using this atomic model we can also demonstrate the reliability of determining Coulomb energies from ΔV_g . For the calculations in Ref. [16] we have $E_c=e^2/\epsilon l_0=7.9$ meV. This value agrees excellently with the determination of the Coulomb energy $E_c=\frac{1}{2}e\Delta V_g=7.5$ meV from the calculated gate-voltage interval in the same paper and demonstrates that this is a reliable and independent method to determine E_c .

Let us now come back to the spectral fine structure of the quantum-dot atoms. At a first glance it seems surprising that the resonance frequency [Figs. 1(a) and 2] depends only slightly on the number of electrons in the atom in contrast to natural atoms. This occurs since in field-effect-confined quantum dots the confining potential has a nearly parabolic shape [16]. In this case the FIR dipole excitation only excites the rigid center-of-mass oscillation of all electrons in the dot at the eigenfrequency ω_0 of the "empty" external potential which is thus independent of N [18,19]. This result is disappointing in the sense that dipole excitation gives no access to the internal fine structure of the quantum-dot atom. For our experiments, however, with a well-defined number of electrons per dot, we observe a clearly resolved frequency shift (see Fig. 2) which oscillates in phase with the number of electrons [Fig. 3(c)]. We find linewidth oscillations and, in a magnetic field, splitting into two reso-

nances. We attribute this behavior to the fact that, due to the Coulomb charging effect, the next electron has to be "squeezed" into the dot. This influences the confining potential in a complex way, in particular, induces deviations from a perfect parabolic shape. These deviations vary in strength as we tune V_g within the interval of a fixed number of electrons. The behavior of half-width and resonance frequency depicted in Figs. 3(b) and 3(c) has been observed consistently on several samples. In particular we observe that the splitting into two resonances is most strongly pronounced in the switching regime. We find that the splitting sets in above a certain magnetic field B_s and that this splitting then remains for all higher $B > B_s$, with the absolute separation between the two peaks decreasing with B , e.g., $\omega_{r2} - \omega_{r1} = 1.6 \text{ cm}^{-1}$ at $B = 3 \text{ T}$ in Fig. 3(a) and it corresponds to 0.6 cm^{-1} at $B = 6 \text{ T}$ in Fig. 1(a). A possible explanation for this splitting is the following. Calculations for a parabolic potential and $N=2$ show that, with increasing B , the ground state switches at a certain B_s to another state with a different angular momentum [17,19]. For a parabolic confinement this cannot be observed in FIR experiments, since the energy difference between the new ground state and the corresponding excited state is still the same. However, recently it has been calculated [20] that the degeneracy of the excited state is lifted for $B > B_s$ if the confining potential is not parabolic. This might account for our observation.

In summary, we have incremented the occupation of field-effect-confined quantum-dot atoms with level separations of 2 meV in discrete steps from 1, 2, 3, to 4 electrons. The discrete number is stabilized by a high Coulomb charging energy of 15 meV and is directly reflected in a steplike FIR absorption strength.

We thank R. Nötzel for the MBE growth of our samples, H. Lage and C. Lange for expert help, and the Bundesministerium für Forschung und Technologie for support.

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