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Excitation of three-dimensional quantum dots

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Arrays of field-effect-confined quantum dots with diameters down to 100 nm have been prepared from $Al_xGa_{1-x}As$-GaAs heterostructures. In far-infrared spectroscopy, transitions are induced between the 2–3-meV separated quantum levels of the lateral confinement. In tilted magnetic fields we observe a resonant coupling to states arising from the confinement in the growth direction. The level spacing in this z direction is found to be only a few meV in our samples and thus comparable to the lateral confinement. We observe in tilted magnetic fields that the excitation spectrum is determined by the *total* magnetic field B. Thus the quantum dots are essentially *three-dimensional* objects.

Low-dimensional quantum-confined electronic systems in semiconductors have recently attracted much interest. The ultimate limit is a quantum dot, an artificial atom, where few electrons are confined in all three dimensions.[1–7] Quantum dots have to date been realized by lateral structuring of two-dimensional (2D) electronic systems. In these systems the lateral confinement of the electrons in the x-y plane is usually assumed to be much smaller than the one in the growth direction z. We will refer to quantum dots in this limit as "quantum-dot disks."

Here we will report on field-effect-confined quantum-dot arrays in GaAs, where both the confinement energy in the z direction and the lateral confinement energy are typically several meV. This leads to interesting coupling effects between states confined in the z direction and the quantum-dot states confined in the lateral directions. In far-infrared (FIR) spectroscopy with perpendicular magnetic fields B, transitions between the quantum levels of the lateral confinement are excited. In tilted magnetic fields, we observe a resonant coupling with states in the growth direction. The resonance frequencies correspond well to those of a three-dimensional harmonic atom, as calculated by Li *et al.*[8] For magnetic fields above the coupling regime the resonance frequencies are determined by the total magnetic field, indicating that the excitation of the quantum dots is three dimensional.

The quantum-dot structures are prepared starting with $Al_{0.32}Ga_{0.68}As$-GaAs heterostructures grown by molecular-beam epitaxy with 2D densities of $N_s = 2 \times 10^{11}$ cm^{-2} and mobilities of $\mu = 900\,000$ cm^2/V s (at 4.2 K). A Si δ-doped layer in the GaAs, deposited at a distance of 330 nm from the $Al_xGa_{1-x}As$-GaAs interface, acts as a back contact to charge the dots. The doping density of 2×10^{12} cm^{-2} was optimized to have enough conductivity for charging the dots but still be semitransparent to FIR radiation. On top of the heterostructure we prepared a periodic photoresist dot array by holographic lithography, with periods a ranging from 500 nm to 200 nm and with photoresist dot sizes of about half the period. An 8-nm-thick semitransparent NiCr gate of 4 mm diameter was evaporated onto the photoresist structure. Outside the gate area the electron gas was removed by etching the $Al_xGa_{1-x}As$. 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.[1,2,4,7] Our back gated structures have another important advantage for the results we obtained. The band bending at the position of the back gate influences the z-confinement potential at the interface and leads to less steep potential curvatures than are usually found in heterostructures. This allows us to create systems with small energy separations in the z direction. FIR transmission spectroscopy was carried out with a Fourier transform spectrometer. With our sample holder the surface normal of the sample could be tilted *in situ* with respect to the magnetic field 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. All experiments were carried out at a fixed temperature, $T = 2.2$ K.

Figure 1 shows experimental FIR transmission spectra for a sample with period of $a = 200$ nm (labeled no. 1 in the following) at fixed magnetic field $B = 9$ T and different tilt angles φ between 0° and 36°. The gate voltage V_g is chosen such that isolated dots with three electrons per dot are formed. The number of electrons is stabilized by a high Coulomb energy of about 15 meV, which we could estimate from the gate voltage interval of the stepwise increasing absorption strength.[7] For $B = 9$ T there is resonant absorption at $\omega_r \approx 125$ cm^{-1} with an amplitude of about 3% (for $\varphi = 0°$). For increasing tilt angle φ the resonance shifts slightly to higher frequencies and broadens; the integrated absorption strength does not vary with φ. It is surprising that the resonance frequency remains nearly constant. For strong z confinement, the quantum-dot disk limit, we would expect that the resonance frequency should decrease because of the smaller in-plane magnetic field. The expected cosine law, indicated by triangles in Fig. 1, does not describe the experiment at all.

Let us first discuss the magnetic field dependence of the resonance frequencies at different tilt angles. We present data of two samples, sample no. 1 from above and a sample with a period $a = 500$ nm, labeled no. 2 in the following. The experimental dispersions at $\varphi = 0°$ is shown as dashed lines in Fig. 2(a). Such a dispersion

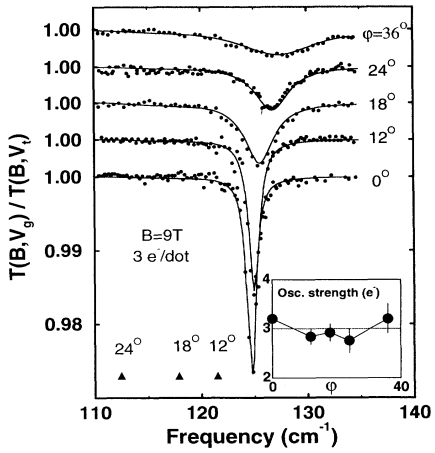

FIG. 1. Experimental FIR transmission spectra (symbols) and fits (solid lines) of quantum-dot sample no. 1 (period a = 200 nm) at B = 9 T and different tilt angles φ from 0° to 36°. The gate voltage V_g is held fixed at a value where isolated dots with three electrons per dot are formed. The inset shows the integrated absorption strength in units of the number of electrons per dot as a function of the angle φ. The integrated absorption strength remains constant within the experimental error. The triangles mark the expected frequency of the resonance with increasing φ for a a quantum-dot disk with strong z confinement (the value of φ = 36° results in an ω_r = 100 cm^{-1}). The spectral resolution is set to 0.25 cm^{-1} and the temperature is T=2.2 K.

has been observed previously.[1,3,4,7] At B = 0 there is a single resonance, ω_r (with ω_r = 11 cm^{-1} for no. 1 and ω_r = 25 cm^{-1} for no. 2) which splits for $B > 0$ into two resonances; one increases with increasing B and approaches the cyclotron frequency $\omega_c = eB/m^*$ and the other decreases with B. This B dispersion can be understood as a one-particle excitation in a two-dimensional parabolic confinement in a magnetic field[9] since the FIR radiation couples only to the rigid center-of-mass motion of all electrons. With regard to the selection rules[1,10] the excitation frequencies are

$$\omega_r^{\pm}(B) = \pm\omega_c/2 + \sqrt{(\omega_c/2)^2 + \omega_r^2}. \qquad (1)$$

The result within the parabolic model describes well the dispersion at φ = 0°. The splitting at small magnetic fields in the few electron system is caused by deviations of the external lateral confinement potential from the parabolic shape.[11]

If we now follow the FIR resonances at a fixed value of φ = 18° in their dependence on magnetic field B, we find the dispersion marked by • in Fig. 2(a). In comparison, Fig. 2(b) shows the result of sample no. 2 with 70 electrons in the quantum dots. For both samples we observe a splitting in the dispersion at final tilt angle, which is clearly resolved in the experimental spectra in Fig. 2(c) for sample no. 2. The splitting is caused by a resonant interaction with states confined in the z direction. It increases with increasing φ and resembles the resonant subband-Landau-level coupling previously observed in tilted-field cyclotron resonance (CR) experi-

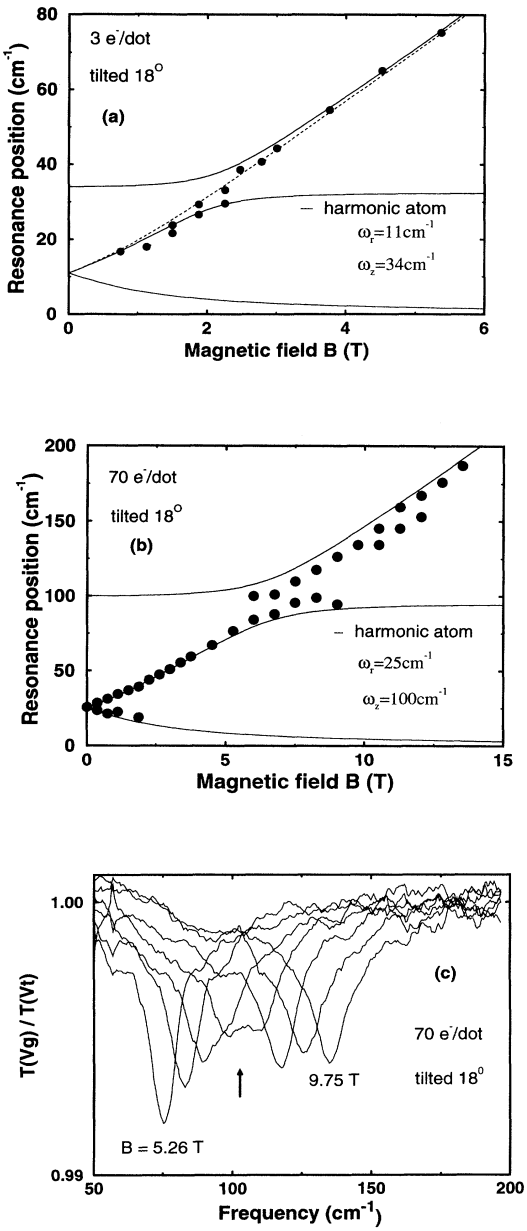

FIG. 2. (a) Magnetic field dispersion of the resonance frequencies (symbols) for sample no. 1 at fixed tilting angle φ = 18°. The solid lines are fits in the model of a harmonic atom with the indicated confinement frequencies ω_r = 11 cm^{-1} in the x-y plane and ω_z = 34 cm^{-1} in the z direction. The dashed lines are the theoretical and experimental results at φ = 0°. (Note that for the ω^- branch the solid and dashed lines are identical within the linewidth.) (b) In comparison, the results of sample no. 2 with a period of a = 500 nm and 70 electrons in the quantum dots are shown. For this sample the lateral confinement energy is ω_r = 25 cm^{-1} and the best fits we obtained with an ω_z = 100 cm^{-1} for the confinement in the z direction. (c) Experimental transmission spectra of sample no. 2 in the anticrossing regime. The interaction with the states in the z direction leads to a splitting of the resonance around B = 7 T.

ments on 2D systems.[12] A similar coupling has been observed in 1D systems[13] and in wide parabolic quantum wells,[14] where the collective character of the resonances is more important, as is also the case in our quantum-dot structures.

To explain the experimentally observed behavior of our quantum dots we need a model which takes into account the confinement in all three directions. A model of a three-dimensional quantum-dot atom, i.e., with parabolic confinement in all three direction, has been proposed by Li *et al.*[8] They calculate the resonance frequencies in the form of a cubic equation in ω^2. If we assume that the shape of potential in the x-y plane has a circular symmetry, this equation reads

$$\omega^6 - \omega^4(\omega_c^2 + 2\omega_r^2 + \omega_z^2) + \omega^2[\omega_c^2(\omega_r^2 \sin^2\varphi + \omega_z^2 \cos^2\varphi) + \omega_r^2(\omega_r^2 + \omega_z^2)] - \omega_r^4\omega_z^2 = 0, \quad (2)$$

where ω_r is the resonance frequency of the lateral confinement, ω_z is the resonance frequency of the confinement in the z direction, and φ is the tilt angle. We have solved this equation numerically and have choosen ω_z to best fit the data. The resonance frequency ω_r and $\varphi = 18°$ are taken from the experiments. The solid lines in Figs. 2(a) and 2(b) mark the results of the fits. For sample no. 1 with $\omega_r = 11$ cm^{-1} we find a resonance frequency in the z direction of $\omega_z = 34$ cm^{-1} and for sample no. 2 with $\omega_r = 25$ cm^{-1} a resonance frequency of $\omega_z = 100$ cm^{-1}. Qualitatively, the model of the parabolic atom describes well the observed frequency dispersions of the quantum dots in tilted magnetic fields. In particular it shows that the resonance frequencies at large fields are governed by the total rather than the perpendicular magnetic field component. Below the anticrossing regime, theory and experiment show that the resonance frequencies follow approximately the cosine-law behavior of a quantum-dot disk. While the lateral confinement is indeed nearly parabolic,[15] this assumption is not very well fulfilled for the z direction in our heterostructure samples. In a good approximation it has a triangular shape. Therefore Eq. (2) cannot determine quantitatively the magnitude $\Delta\omega$ of the splitting at the resonance position with the states in the z direction. The structure in Fig. 2(b) at about $\omega = 150$ cm^{-1} is most probably caused by a resonant interaction with the next higher state in the z direction. Such an interaction does not occur for the special case of parabolic potentials in all three directions as assumed in Eq. (2). Nevertheless, in general the applied model explains well all important features of the experiments. From this we conclude that the excitation of our quantum dots is indeed *three* dimensional. The quantum dots undergo a transition from quantum-dot disks to three-dimensional quantum dots at the anticrossing regime. Transitions to a three-dimensional behavior have been investigated theoretically[16] and found in tunneling experiments on wide parabolic quantum wells,[17] in CR experiments on inversion layers in InSb,[18] and in luminescence studies on 1D systems.[19]

Let us now discuss an additional feature observed in Fig. 1. The resonance linewidth broadens with increasing φ. We believe that this is another intrinsic feature of the system which is not expected at first sight. CR resonance studies on 2D-electron systems show that the CR linewidth is smaller at $\varphi = 90°$.[20] If, as in our experiments, a three-dimensional motion of electrons is possible then the resonance frequency depends on the center coordinate of the cyclotronlike motion in the quantum dots. In 2D systems this was extensively studied and leads to two possible excitations in the tilted field experiments, a "three-dimensional" CR for electrons that are repelled by the Lorentz force from the interface and diamagnetically shifted intersubband resonances for electrons that execute skipping orbits at the interface.[21] For a quantum dot with confinement in all three dimensions this should result, depending on the shape of the z potential, in a number of resonance frequencies that appear as a broadening in the experiments.

In summary we have presented FIR transmission experiments on quantum dots in tilted magnetic fields. The quantum-dot systems show a resonant interaction with states confined in the growth direction from which we can determine the level spacing in the z direction and which turns out to be comparable with the one in the lateral direction. Thus we can excite three-dimensional motion in the quantum dots which is manifested in the observation that the dispersion of the resonance frequencies at large magnetic fields depend on the total rather than on the perpendicular component of the magnetic field.

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