Nonlocal Dynamic Response and Level Crossings in Quantum-Dot Structures

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Very small quantum-dot structures containing 210 to 25 electrons per dot have been prepared starting from modulation-doped AlGaAs/GaAs heterostructures. The far-infrared response consists of a set of resonances which split, in a magnetic field B, into branches with negative and positive B dispersion. The intersection of these resonances, in classical analogy edge magnetoplasmons, leads to an anticrossing of the dispersions. This resonant coupling is induced by nonlocal interaction which becomes important at small dimensions.

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Currently there is an increasing interest in the investigation of ultrasmall, laterally microstructured, originally two-dimensional electronic systems (2DES). Because of the reduced dimensionality, quantum confinement and novel transport phenomena are observed.[1-6] One ultimate goal is the realization of artificial atoms in quantum-dot structures. We have prepared quantum-dot structures containing 210 to 25 electrons per dot in AlGaAs/GaAs heterostructures by deep-mesa-etching techniques.[7] The dynamic far-infrared (FIR) response shows a set of resonance absorption peaks at frequencies ω_{0i} which split with increasing magnetic field B into two branches, $\omega_{i+}(B)$ and $\omega_{i-}(B)$, with positive and negative B dispersion, respectively ($i=1,2,\ldots$). The dispersion is similar to that observed for excitations in InSb quantum-dot structures[6] and for edge magnetoplasmons in wider (radius $R=1.5$ μm) GaAs structures[8] and finite-sized 2DES on liquid He surfaces (Refs. 9 and 10; for a recent review, see, e.g., Ref. 11). However, in our systems we can for the first time observe higher-order branches ($i=2$) in ultrasmall dimensions ($R\approx 10a_0^*$, a_0^* the effective Bohr radius). For these small structures we find, very surprisingly, a resonant anticrossing of the ω_{1+} and the ω_{2-} branches.

We show that this anticrossing arises from nonlocal interaction. We find that the coupling is similar in strength, but very different in the ω vs B dependence, as compared to resonant nonlocal interaction in infinite 2DES.[12] The nonlocal interaction is governed by the parameter a_0^*q ($q=i/R$ is the wave vector, see below). Thus the smallness of our structures makes it possible to observe the interaction which is, as such, an inherent property of quantum-dot structures. For a system with a very small number of electrons, which behaves more single-particle-like, we expect that the observed anticrossing corresponds to transitions of anticrossing single-particle energy states in a magnetic field, i.e., the bifurcation that has been observed indirectly in magnetocapacitance measurements on quantum-dot structures.[5] The anticrossing that we observe is a demonstration of electron-electron interaction in the artificial atom in the sense that it is sensitive to a fine structure in the excitation spectrum beyond non-self-consistent "empty-atom" models, e.g., Ref. 13.

The samples were prepared by starting from modulation-doped AlGaAs/GaAs heterostructures. An array of photoresist dots (with a period of 1000 nm both in the x and in y directions) was prepared by a holographic double exposure. Using an anisotropic plasma-etching process rectangular 200-nm-deep grooves were etched all the way through the 10-nm-thick GaAs cap layer, the 53-nm-thick Si-doped AlGaAs layer, and the 23-nm-thick undoped AlGaAs spacer layer into the active GaAs, leaving quadratic dots with rounded corners and geometrical dimensions of about 600 by 600 nm (see inset of Fig. 1). With this technique of "deep mesa etching" it was possible to realize in linear stripe systems 1DES with typical energy separation for the 1D subbands of about 2 meV.[7,14] It was found that the actual width of the electron channels was smaller than the geometrical width, indicating a lateral edge depletion of 100 to 120 nm. For the dot structures here, with increased etched surface area, this depletion is even more pronounced. Actually, we have prepared samples which had, in the dark, no mobile electrons. Via the persistent photoeffect we could then increase the number of electrons in steps up to 210 per dot. Since lateral transport[1] is inherently not possible in dot structures, we used the strength of the FIR absorption to determine the number of electrons per dot, N. In particular we used the Drude-type model for bound electrons in Ref. 15, formula (6). The potential that confines the electrons and thus determines the radius of the 2D disk, depends on the remote ionized donors and, in a self-consistent way, on N. We have estimated the radius from the observed resonance frequency and formula (1) which will be explained below.

The FIR experiments were performed in a superconducting magnet cryostat, which was connected via a waveguide system to a Fourier-transform spectrometer. The transmission $T(B)$ of unpolarized FIR radiation through the sample was measured at fixed magnetic

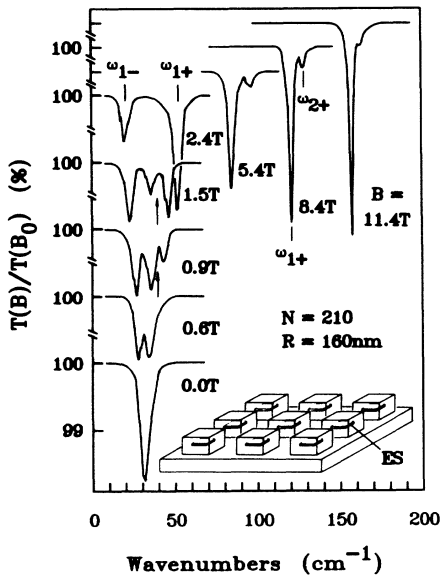

FIG. 1. Relative transmission of unpolarized FIR radiation for a quantum-dot structure with radius $R=160$ nm and $N=210$ electrons per dot. In a magnetic field $B>0$ the resonance splits into two branches, ω_{1+} and ω_{1-}, with positive and negative B dispersion, respectively. For $B>4T$ an additional resonance, ω_{2+}, is observed. At $\omega\approx 40$ cm^{-1} (↑) there is a pronounced anticrossing intersection. Inset: The dot structure shown schematically. The actual dots have rounded corners. ES denotes the confined electron system.

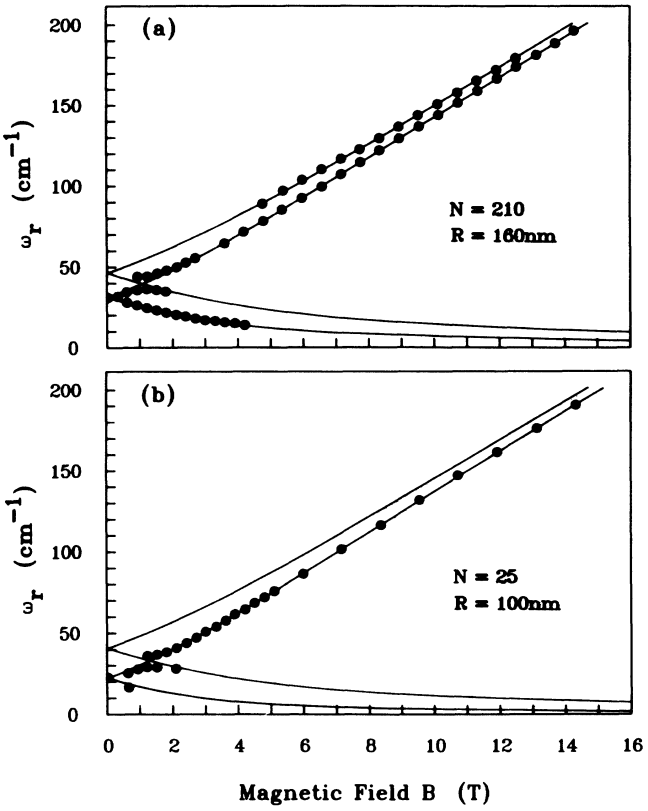

FIG. 2. Experimental B dispersion of resonant absorption in quantum-dot structures with (a) $R=160$ nm and $N=210$ and (b) $R=100$ nm and $N=25$. The full lines are fits with the theoretical dispersion [Eq. (2)]. Both structures show an anticrossing of the ω_{2-} with the ω_{1+} mode, which is caused by nonlocal interaction.

fields, B, oriented normally to the surface of the sample. The spectra were normalized to a spectrum $T(B_0)$ with a flat response. The resolution of the spectrometer was set to 0.5 cm^{-1}. The temperature was 2.2 K. The measured sample area was 3×2 mm^2; thus we measure 10^7 dots.

Experimental spectra for a sample with a dot radius $R=160$ nm and $N=210$ electrons per dot are shown in Fig. 1. For $B=0$ one resonance is observed at $\omega_0=32$ cm^{-1}. With increasing B the resonance splits into two resonances; one, ω_{1-}, decreases in frequency, the other ω_{1+}, increases. For $B>4T$ a second resonance, ω_{2+}, can be resolved which also increases with B. The most interesting observation is the obvious resonant coupling which occurs at a frequency of about 40 cm^{-1}. The experimental resonance positions for this sample are depicted in Fig. 2(a). Figure 2(b) shows the resonance positions from a sample containing only 25 electrons per dot. The dip in $T(B)/T(B_0)$ is only 0.4% which is the experimental limit for reliably determining position and signal strengths for the current spacings of the dots. Here we cannot clearly resolve the ω_{1-} branch because of the limited sensitivity of our spectrometer at small frequencies. Similar dispersions as in Fig. 2(a) were measured on a series of samples with slightly different values of R, N, and corresponding ω_0. The interesting observation is the significant resonant anticrossing at $\omega\approx 1.4\omega_0$, which was found for all our samples.

In the following we will discuss that this anticrossing is caused by nonlocal interaction. For our discussion we first adopt a description for a classical 2DES of finite size and discuss effects of quantum confinement later. The FIR resonances observed here, are, except for the resonant splitting, very similar to earlier observations on larger, finite-sized 2DES in GaAs (dots with $R=1.5$ μm)[8] and electrons-on-liquid-He systems ($R\approx 1$ cm)[9,10]. These resonances can be explained either in an edge-magnetoplasmon model[9-11] or, at least at small B, equivalently as a depolarization resonance.[8] A simple way to describe the FIR response of a finite-sized 2DES is to start from linear edge magnetoplasmons (e.g., see Refs. 11, 16, and 17) which have the dispersion $\omega_{ep}^2=0.81\omega_p^2(q)$, where $\omega_p^2(q)=N_s e^2 q/2m^*\epsilon_0\epsilon_{\rm eff}$ is the 2D plasmon frequency. The circumference of the disk quantizes the q vectors in values $q=i/R$ ($i=1,2,\ldots$). Thus that at $B=0$ is

$$\omega_{0i}^2=0.18N_s e^2 i/2m^*\epsilon_0\epsilon_{\rm eff}R\,. \qquad (1)$$

In a magnetic field one calculates a set of double

branches (e.g., see Refs. 8 and 10),

$$\omega_{i\pm} = [a_{i1}\omega_{0i}^2 + a_{i2}(\omega_c/2)^2]^{1/2} \pm a_{i3}\omega_c/2 , \quad (2)$$

with $a_{i1} = a_{i2} = a_{i3} = 1$ (ω is the cyclotron frequency). These models already agree very well with more sophisticated theories which determine more accurately the coefficients a_{ik} ($k = 1,2,3$) of ω_{0i} and ω_c. The latter and the spacings of higher modes depend slightly on the exact modeling of the 3D density profile for the electron distribution.

We have fitted in Fig. 2(a) the dispersion with formula (2), using a_{21} as a fitting parameter and all other $a_{ik} = 1$, and find, taking account of the nonparabolicity of m^* in GaAs, a very good agreement with the experimental dispersion. In particular we find $\omega_{02} \approx 1.5\omega_{01}$, which is very close to the simple-model value of $\omega_{02} = \sqrt{2}\omega_{01}$. From this fit it becomes clear that the splitting is caused by an anticrossing of the ω_{2-} and ω_{1+} modes. The interesting question is which interaction causes this splitting and what determines its strength. The fact that this splitting is not observed on larger GaAs systems with a very similar shape,[8] in particular, not on the liquid-He systems of Ref. 9, where beautifully sharp intersecting edge magnetoplasmons were found with, however, no interaction at all, leads to the conclusion that the smallness of our structures is the important parameter. (For large R and thus small q, ω_{i+} and ω_{i-}, represent, respectively, left- and right-circular-polarized eigenmodes, which are as such decoupled.) This automatically draws one's attention to the nonlocal interaction. Nonlocal effects are well known for the homogeneous 2DES. They arise from the inherent finite compressibility of the Fermi gas and lead to corrections $q^2 v_F^2$ (v_F = Fermi velocity) for the squared plasmon frequency $\omega_p^2(q)$.[12,17] These effects are very small under usual experimental conditions. However, they can be clearly observed in a resonant-magnetic-field experiment, where the nonlocal interaction leads to an anticrossing with $2\omega_c$.[12] That is, the "local" magnetoplasmon dispersion, $\omega_{mp}^2 = \omega_{p0}^2 + \omega_c^2$, splits into two branches. One branch starts at $B = 0$ with the local dispersion at ω_{p0} and with increasing B approaches asymptotically $2\omega_c$. The other branch starts at $\omega = 0$ asymptotically with $2\omega_c$ and with increasing B approaches the local dispersion. Thus, the frequency dependence of the nonlocal interaction in 2D is very different from the dot systems shown in Fig. 2.

There is so far no theoretical treatment of resonant nonlocal interaction in 0D system. Only nonresonant nonlocal effects on the linear 1D edge-magnetoplasmon dispersion have been considered until now.[17] Nevertheless it is interesting to compare the 2D nonlocal interaction strength with the splitting that is observed here, in particular, since the splitting occurs at small B, and thus one is not too far away from a 2D-plasmon-type behavior. The strength of the nonlocal resonant coupling in 2D, measured in terms of the frequency splitting,[12] is $\Delta\omega/\omega_0 = 2.6\sqrt{q}$ (q in units of nm^{-1}). If we use for the interaction here the same model and for the ω_{2-} branch $q = 2/R$, we find for the sample of Fig. 2(b) with $R = 100$ nm $\Delta\omega/\omega_0 = 0.37$ which agrees surprisingly well with the experimental value of 0.33. This very close agreement might be to a certain degree accidental and should be compared with a so far not available rigorous theory. In particular, we expect that the nonlocal interaction also depends on the exact three-dimensional density profile of the electron system. However, this close agreement demonstrates that the experimentally observed splitting is of the expected order for nonlocal interaction. Moreover, independently of the absolute value we expect within our simple analogy $\Delta\omega/\omega_0 \propto (1/R)^{1/2}$. If we scale the experimental splitting of 0.33 for the sample in Fig. 2(b) ($R_1 = 100$ nm) with $(R_1/R_2)^{1/2}$ we find 0.26, which agrees very well with the experimental splitting of 0.25 for the sample in Fig. 2(a) ($R_2 = 160$ nm). (For all our samples with R ranging from 100 to 180 nm we found that the experimental splitting agrees within a factor of 0.85 to 1.0 with $\Delta\omega/\omega_0 = 2.6\sqrt{q}$.) Thus also the dimensionality dependence confirms that the interaction is caused by nonlocality. The question arises whether deviations from a circular shape might influence the splitting. We do not believe that this is a significant effect: (i) Such effects are independent of the dimensions and should thus also be present in the experiments of Ref. 8 where samples of a very similar shape show no splitting. (ii) For our samples one would expect that a splitting due to the geometrical shape effect should be especially pronounced at large N and R when the electronic system extends closer to the geometrical edge. This is in contrast to the experimental observation.

The smallness of the structures, needed to make nonlocal effects essential, leads inherently to the regime of quantum confinement. Concerning our structures we note that linear stripes with the same dimensions, prepared with the same techniques as used here, show a quantum-confined 1D energy spectrum with a typical subband separation of about $\hbar\Omega_0 = 2$ meV.[7] Therefore we believe that, in particular, in our sample with only 25 electrons per dot, the electrons occupy discrete quantized energy levels. In particular, in a two-dimensional harmonic-oscillator potential,[13] $V(x,y) = \frac{1}{2} m^* \Omega_0^2 (x^2 + y^2)$, only five discrete energy levels are occupied. It is well known (e.g., see Refs. 14 and 18) that the level spacing $\hbar\Omega_0$ is not directly observed in a FIR experiment, but rather the observed resonance frequency ω_r is shifted, $\omega_r^2 = \Omega_0^2 + \omega_p^2$, where ω_p characterizes the collective depolarization effect which increases with increasing N. For the sample with $N = 210$ the resonance frequency $\hbar\omega_r = 4$ meV is strongly governed by the depolarization effect. Thus here a classical, plasmonlike resonance behavior is the adequate description of the FIR response. However, for the sample with $N = 25$ electrons per dot we have estimated $\hbar\Omega_0 = 2$ meV; thus ω_p is about equal

to or smaller than the one-particle energy separation Ω_0. Thus here the FIR response should reflect strongly the single-particle aspects of the dot structure.

The collective effects make it difficult to determine exactly the level spacing of a 0D system from FIR spectroscopy. In particular, from the experimental B dispersion only, one cannot distinguish between a classical collective plasmon type of response [Ref. 8 or formula (2) here] and 0D-level transitions, e.g., in a harmonic-oscillator confinement.[6,13] With a theoretical modeling of the splitting that we observe here, one can perhaps gain insight into the energy structures of the dots. As was pointed our for linear systems[11] the edge magnetoplasmons correspond in a quantum-confined system to transitions between the discrete energy states. As such, in the limit of quantum confinement, and for the $N=25$ sample with $\omega_p < \Omega_0$, we approach this limit, the observed splitting corresponding to transitions between anticrossing one-particle energy levels in a magnetic field.

In conclusion, in the FIR response of quantum-dot structures containing 25 to 210 electrons per dot a resonant anticrossing of edge-magnetoplasmon-type excitations is observed. The interaction arises from the nonlocality which becomes important for the very small dimensions. The interaction is, compared to 2DES, similar in strength, but very different in its ω vs B dependence.

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