Coupling between LO phonons and electronic excitations of quantum dots

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The far-infrared response of self-assembled $In_xGa_{1-x}As$ quantum dots located a distance from a two-dimensional $Al_xGa_{1-x}As/GaAs$ interface has been studied as a function of magnetic field. An avoided crossing at about 45 meV, which involves the lowest electronic transition of the dots, has been observed. Calculations of the interaction between the excitations of these quantum dots and the LO phonons of the $Al_xGa_{1-x}As/GaAs$ interface have been made, and the resulting splitting has been found to be in agreement with the experimental results. The calculated dependence of the magnitude of the splitting on the separation between the dots and the nearest interface is consistent with the experimental results. [S0163-1829(97)01327-1]

I. INTRODUCTION

Studies of the optical properties of quantum dots related to their confined electronic states have been attracting considerable attention recently.[1–3] Dots formed by so-called coherent island growth[3–7] are particularly attractive because of their small, uniform sizes, their large quantization energies, and their high-quality interfaces. Improvements in the quality of samples and in the understanding of their electronic properties recently have made possible quantitative studies of carrier-phonon interactions in these systems, and some work along these lines is beginning to be reported.[8] Carrier-phonon interactions not only are essential to understanding the optical properties of such systems, but they also control such phenomena as carrier relaxation, which is of particular interest in quantum dots.

Carrier-phonon interactions have been central to the study of semiconductors for many years, and the interactions of electronic transitions with LO phonons in magneto-optical experiments have been a powerful spectroscopic way of studying them. In the weak-coupling regime carrier-phonon interactions give rise to polaron mass enhancements and carrier relaxation. When an electronic excitation energy becomes comparable to a LO-phonon energy, strong coupling between them can occur. For example, it is known in the bulk that this interaction can lead to LO phonons being effectively bound at shallow donors.[9]

In studies of LO phonons in quantum wells and other nanostructures[10] it has been found that the LO-phonon spectrum consists of so-called confined modes, which exist in only one of the different layers, and interface modes, which are associated with the interfaces. In a recent work[11] Knipp, Pierson, and Reinecke have studied the coupled modes arising from the electronic excitations and the LO phonons of spherical quantum dots. They found that when these energies become comparable, hybrids involving both the confined phonons and also the interface phonons coupled with the electronic excitations result, and they strongly affect the optical properties of the quantum dots for energies in the neighborhoods of these crossings.

In the present work we have studied in magnetotransmission the electronic excitations of quantum dots formed by self-organized growth of $In_xGa_{1-x}As$, which are embedded in a suitable GaAs field-effect-transistor structure of the type discussed in, e.g., Refs. 3 and 12. In these structures the quantum dots are separated from two-dimensional $Ga_{1-x}Al_xAs$ or AlAs interfaces by varying distances. For cases in which the interfaces are sufficiently near the dots we observe an anticrossing involving an electronic excitation and another excitation. We have made calculations of the hybridization of the electronic transitions and the LO phonons of an interface separated by a distance from the dots, and we find that the resulting splitting agrees well with the experiment. From these results we interpret the observed resonances as hybridizations of a transition between the two lowest electronic states of the dots and the interface phonons. Our model allows us to investigate the influence of the separation between the dot and the interface on these couplings. We find that the theoretical results are in accord with experiment for this dependence.

II. EXPERIMENT

The heterostructures with embedded self-assembled $In_xGa_{1-x}As$ dots are grown by molecular-beam epitaxy on semi-insulating GaAs substrates. The layer sequence before

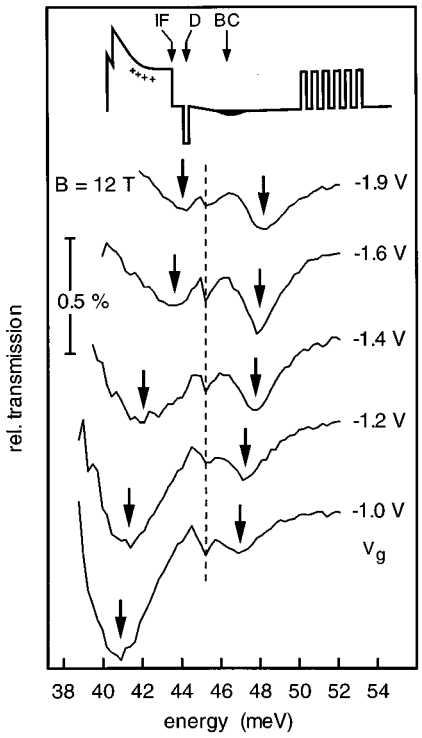

FIG. 1. Normalized transmission spectra of self-assembled $In_xGa_{1-x}As$ quantum dots at $B=12$ T for several gate voltages V_g. As V_g is decreased, the dots become more and more depleted and the total oscillator strength decreases. At the same time the resonances shift to higher energies and an exchange of relative oscillator strength is observed. The dashed line indicates the energy of the AlAs-like interface phonons. Inset: schematic representation of the conduction band. Arrows indicate the positions of the interface (IF), the $In_xGa_{1-x}As$ dots (D), and the back contact (BC).

the deposition of the dots is composed of a smoothing superlattice (30 periods of 3 nm of GaAs and 3 nm of $Al_{0.3}Ga_{0.7}As$), a 1 μm GaAs buffer, a back contact layer that is Si δ doped (1×10^{12} cm^{-2}), and a 97 nm GaAs spacer layer. For the deposition of the dots, the substrate temperature is lowered from 580 °C to 530 °C and cycles of 0.5 monolayers of $In_{0.5}Ga_{0.5}As$ with 3 s of growth interruption (with As flux on) are repeated while the surface morphology is monitored by reflection high-energy electron diffraction (RHEED). When the RHEED pattern turns spotty, which indicates the transformation from two- to three-dimensional (Stranski-Krastanow) growth, the $In_xGa_{1-x}As$ deposition is stopped and the resulting dots are capped with 3 nm of GaAs. After raising the temperature to 600 °C, a 15 nm $Al_{0.3}Ga_{0.7}As$ spacer is grown. Between the last two layers is the $Al_xGa_{1-x}As$/GaAs interface of interest. The layer structure is completed by 40 nm of Si-doped $Al_{0.3}Ga_{0.7}As$ ($n=2.3\times10^{18}$ cm^{-3}), 50 nm undoped $Al_{0.3}Ga_{0.7}As$ and a 10 nm GaAs cap. Further details of the growth procedure and the morphology of the dots can be found in Refs. 4 and 13. We estimate that the diameter and the height of the dots are 25 and 6 nm, respectively. Samples of 5×5 mm^2 are cleaved from the wafer and wedged to avoid Fabry-Pérot interference. Small In pellets are alloyed to provide Ohmic contacts to the back contact layer (BC in Fig. 1). Evaporation of a 5-nm-thick semitransparent NiCr gate completes the sample

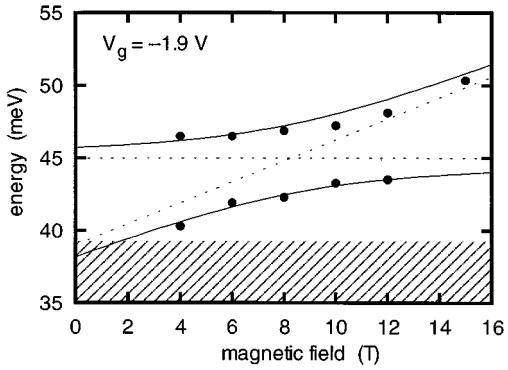

FIG. 2. Measured (circles) and calculated (solid lines) resonance positions in the region where coupling between the dot resonances and the interface phonons occurs. Dotted lines give the theoretical resonance positions without coupling. In the vicinity of the reststrahl band (shaded area), the signal-to-noise ratio decreases dramatically, so that the experimental resonance positions can no longer be determined.

preparation. By application of a suitable gate bias between the semitransparent top gate and the back contact, the $In_xGa_{1-x}As$ dots can be loaded controllably with electrons.[3,12] The inset in Fig. 1 displays a schematic representation of the conduction band in the sample for a gate bias just below depletion of the dots.

All measurements are carried out at liquid-He temperatures. The samples are mounted in the center of a superconducting solenoid, which allows magnetic fields of up to $B=15$ T to be applied perpendicular to the plane of the dots. Using a rapid-scan Fourier transform spectrometer, the far-infrared transmission is recorded in the energy range between 5 and 100 meV. To eliminate spectral features arising from the optical components in the setup, the spectra are normalized by a reference measurement, which is taken at a gate voltage where no electrons occupy the dots or at a magnetic field where all resonances are shifted away from the energy range of interest.

Figure 1 shows the relative transmission spectra of the dots for several gate voltages V_g at a magnetic field of $B=12$ T. Three features are observed in the data: a low-lying resonance, which has a strong upward shift and a strong decrease in strength with increasing negative bias, a higher resonance, which has only a small upward shift and small increase in strength with bias, and a peak at 45 meV, which is not affected much by changing the bias (dashed line in Fig. 1). The 45-meV resonance is similar in energy to the AlAs-like interface phonons of $Ga_{1-x}Al_xAs$/GaAs interfaces, and we attribute it to an AlAs-like mode of the $Al_xGa_{1-x}As$/GaAs interface, which is in agreement with, for example, Ref. 14. The first two resonances (arrows) show an anticrossing behavior with the typical exchange in relative oscillator strength as a function of bias. For these resonances the total oscillator strength decreases with decreasing bias because the number of electrons per dot decreases.

Similar behavior is observed when the energy of the dot resonances are shifted through the interface phonon energy by a variation of the magnetic field. The full circles in Fig. 2 give the measured resonance positions as a function of B at $V_g=-1.9$ V. An anticrossing is observed around $B=8$ T at

an energy of about 45 meV, with a splitting of approximately 4.4 meV between the two lines. This is the splitting that is discussed in the following.

III. THEORY

The dots are taken to have cylindrical symmetry about the growth axis, which implies that the angular momentum m around this axis is a good quantum number. The electronic states of the dots are taken to involve only the lowest quantized state for motion along the growth axis and to be two-dimensional harmonic-oscillator states for motion in the perpendicular plane. These states have been shown to give a reasonable representation of the optical properties of these dots.[3,12,15] For the states with $m=0$ and ± 1, the wave functions in the plane are

$$\psi_m(r,\varphi)=\left(\frac{1}{\sqrt{\pi(L^2/2)^{m+1}m!}}\right)r^{|m|}e^{-r^2/L^2}e^{im\varphi}, \qquad (1)$$

where the characteristic length is $L=(2\hbar/m_e\omega_0)^{1/2}$, ω_0 is the harmonic-oscillator frequency of the effective two-dimensional potential, m_e is the electron effective mass, and r,ϕ are the magnitude of the radial coordinate in the plane and the angular coordinate. The lowest-lying electronic states with $m=0$ and ± 1 are often called the s and p-states of these dots. In a perpendicular magnetic field B the transition energy between the lowest (s) and next higher (p) states is given by

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

where the cyclotron frequency is $\omega_c=eB/m_e$ and $\hbar\omega_0=E_1-E_0$ at $B=0$.

The frequencies of the bulklike LO phonons of the GaAs and the $In_xGa_{1-x}As$ are too low to account for the mode at $\sim$45 meV. Furthermore, the carriers do not interact with the confined phonons of the substrate side superlattice because the superlattice is physically separated from dots and the potentials of the confined phonons are nonzero only within the superlattice layers.[16,17] Therefore, we will be concerned with the coupling of the excitations of the quantum dot with the phonons of the closest $Al_xGa_{1-x}As$ interface, labeled IF in Fig. 1. For simplicity we will model the phonons by the interface phonons of a single GaAs/AlAs interface a distance h away from the plane containing the dot. These modes result from the interface polarization charge of the GaAs/AlAs interface and are obtained from the standard condition that $\nabla\cdot\mathbf{D}=0$ and electrostatic boundary conditions.[16] The interaction between electronic charges and the interface phonon modes is via the electrostatic Fröhlich interaction.[18] The potentials of the interface phonons decay exponentially with distance from the interface, and for a single interface the phonon energy does not depend on the wave vector parallel to the interface.

In this case, the energies of the coupled electronic transition and the interface phonons can be obtained analytically and are given in a manner similar to that in our previous work[11,19] by

$$E=\hbar\omega_{\mathrm{IF}}-I\left(\frac{2}{E-\Delta E-2\hbar\omega_{\mathrm{IF}}}+\frac{1}{E-\Delta E}\right), \qquad (3)$$

where I is the coupling factor

$$I=(0.32e^2\ \ \mathrm{meV})\int_0^\infty dq\ e^{-2hq}|M(q)|^2 \qquad (4a)$$

and

$$M(q)=2\pi\int_0^\infty r\ dr\ \psi_0(r)\psi_1(r)J_1(qr), \qquad (4b)$$

where h is the distance between the dots and the interface. Here $\hbar\omega_{\mathrm{IF}}$ is the energy of the interface phonons, ΔE is the electronic excitation energy in Eq. (2), e is the charge of the electron in Gaussian units, and q is the wave vector of the interface mode that is summed over here. Note that planar interface phonons with all q's contribute to the coupled state in Eq. (4).

IV. DISCUSSION

In the present experiments, we have studied several samples. In some cases the nearest $Al_xGa_{1-x}As$ interface was grown about 30 nm from the layer containing the quantum dots and in others it was grown about 3 nm from this layer. In addition, by varying the gate voltage on the field-effect structure, the electronic transition energies can be changed by modest amounts, which probably results from changes in the screening behavior of the highly doped back contact. For separations of 30 nm between the dot and the interface, avoided crossings like those shown in Figs. 1 and 2 could not be detected.[12] For 3-nm separations, such anticrossings are always seen and their positions vary somewhat depending on the bias conditions (cf. Fig. 1).

The case shown in Fig. 2, in which the gate voltage is -1.9 V, is typical of our results. From transmission studies of similar quantum dots, we know that the wave function in Eq. (1), which corresponds to a two-dimensional harmonic potential, gives a good representation of the transition energies ΔE and that the data can be described well using an effective mass $m_e\approx 0.07m_0$.[3,12,20] Because of the strong absorption in the reststrahl regime, which is indicated by the shaded region in Fig. 2, we are not able to determine directly the effective oscillator frequency for the two-dimensional potential ω_0 for $V_g=-1.9$ V. However, from a careful analysis of data taken at different gate voltages and at different magnetic fields we can extrapolate the value $\hbar\omega_0\approx 39$ meV, which was used in the calculations here.

We have taken the frequency of the interface phonon energy to be 45 meV from Fig. 1 and from Ref. 14. This corresponds to the AlAs-like interface phonons of a single GaAs/AlAs interface. In the evaluations of the coupled modes, we find that the magnetic-field dependence arises mainly from the energies of electronic states, and we neglect the field dependence of the interaction integral I. Thus we use the expression in Eq. (2) for the uncoupled electronic transition energies and the field independent form in Eq. (1) for the wave functions. Then the dotted lines in Fig. 2 give the energies of the uncoupled interface phonons and the elec-

tronic excitation ΔE, and the solid lines give the energies of the coupled modes from our calculations. From our calculations the interaction integral in Eq. (4) is 5.7 meV^2, and we find a splitting of 4.75 meV at the avoided crossing. This splitting is in good agreement with the experimental results of 4.4 meV from the data shown in Fig. 2.

We find that the magnitude of the splitting between the coupled modes falls off approximately exponentially with distance. This dependence can be seen from Eq. (4). For example, for a separation of 30 nm between the dots and the interface, the splitting is only 0.33 meV. This result is consistent with the experimental finding that in samples with such large separations no avoided crossing can be observed.

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