Terahertz-frequency electronic coupling in vertically coupled quantum dots

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We have studied terahertz absorption of samples containing two layers of self-aligned, self-assembled InAs quantum dots separated by a thin GaAs barrier. The vertically coupled dots were charged with electrons by applying a voltage bias between a metal gate and a doped layer beneath the dots. For a positive gate bias corresponding to flatband conditions, an absorption peak was observed near 10 meV (2.4 THz). The absorption is attributed to the inhomogeneously broadened transition between the quantum mechanically split levels (bonding and antibonding states) in the vertically coupled quantum dots. © *2000 American Institute of Physics.* [S0003-6951(00)04252-2]

Self-assembled semiconductor quantum dots are three-dimensionally confined nanostructures which have been studied extensively in recent years. The optoelectronic properties of quantum dots can be tailored by adjusting their size, shape, and composition during growth. However, the tuning of energy levels in quantum dots is limited by the kinetics and thermodynamic processes which govern the growth. This limitation is not present in quantum wells, where wave functions can be engineered by stacking quantum wells with different thicknesses and compositions. Electronic coupling of quantum dots, i.e., growth of artificial quantum dot molecules, represents a major step in tailoring the electronic properties of nanostructures.[1] Besides greater flexibility, the electronic coupling between quantum dots allows one to shift optical transitions to a specific spectral range, like the terahertz (THz) spectral range. Coupled quantum dots with THz-frequency transitions have been proposed as quantum bits in a semiconductor-based quantum computer.[2]

Vertically coupled quantum dots can be obtained by different means. Starting from coupled two-dimensional heterostructures, lateral confinement can be achieved by etching using standard nanolithography techniques, or by depositing a metallic gate on top of a heterostructure. Vertical electronic coupling between quantum dots has been observed experimentally by conductance measurements.[3] Another route to electronic coupling relies on stacking self-assembled quantum dots. Electronic coupling, mainly governed by the barrier separation thickness, is allowed due to the strain-field-assisted vertical self-organization of the quantum dots. Electronic coupling between stacked quantum dots has already been observed by photoluminescence spectroscopy,[4,5] and investigated theoretically.[6,7] However, no direct measurement of the absorption between the quantum mechanically split bonding and antibonding states of the coupled dots has been reported. The associated infrared absorption would correspond in this case to an intraband transition between the energy levels of vertically coupled dots, active for THz electric field polarized in the growth direction. For electric field polarized perpendicular to the growth direction, intraband absorption has been reported in standard self-assembled quantum dots at energies above 40 meV.[8,9]

The investigated samples were grown by molecular beam epitaxy. A typical structure is shown in Fig. 1. The field-effect structure consisted of a GaAs *n*-doped backcontact, an undoped GaAs layer, an AlAs/GaAs blocking barrier, and a GaAs cap layer. The quantum dot carrier population was controlled by an Al Schottky barrier. Au/Ge/Ni was evaporated and alloyed to contact the *n*-doped back layer. The quantum dot molecules were grown in the undoped GaAs layer.[8] Three different types of InAs/GaAs molecules were investigated: ''asymmetric'' molecules (sample A, 7 nm GaAs barrier thickness)[10] where the quantum dot vertical sizes were intentionally different: ≈1.5 nm height for the first quantum dot layer and ≈6 nm height for the second dot layer; and symmetric quantum dot molecules (samples B and C) where the dots were similar (6 nm height) and separated by a GaAs barrier with two different thicknesses (7 and 7.5 nm for samples B and C, respectively).

The quantum dots are lens shaped with a typical diameter of 20 nm. Under the growth conditions, a significant intermixing between In and Ga is expected to occur, thus leading to the formation of InGaAs quantum dots. For infra-

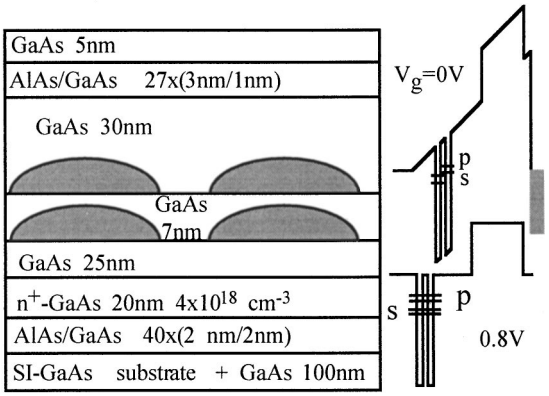

FIG. 1. Schematic structure of sample B. The inset shows the conduction band edge profile under two different applied gate voltages (0 and 0.8 V, respectively).

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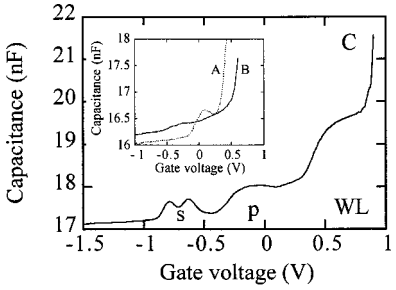

FIG. 2. Capacitance of sample C as a function of the applied gate voltage. The capacitance is measured at 4.2 K. The inset shows the capacitance of sample A (dashed line) and B (full line). The capacitance was measured at 34 Hz with a 5 mV tickle amplitude. The successive loading of *s*,*p*, and wetting layer states is indicated.

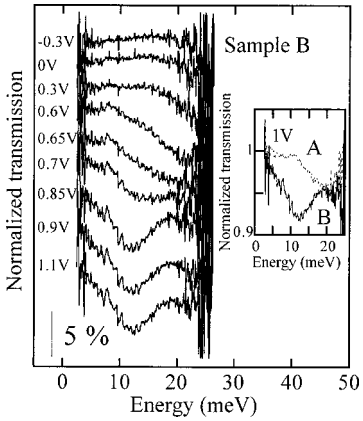

FIG. 3. Normalized far-infrared transmission of sample B as a function of the applied gate voltage. The curves have been offset for clarity. The inset shows a comparison between the normalized transmission of sample B (full line) and the transmission of sample A (dotted line) (1 V gate voltage).

red measurements, an Al metal film was evaporated on the backside of the sample to form an infrared wave guide. Using a Fourier transform infrared spectrometer, far-infrared light was injected through the cleaved edge of a 7 mm long sample.[11] The sample was mounted in a liquid helium cooled cryostat in front of a 1.8 K bolometer.

The capacitance between the Schottky contact and the n^+-buried backcontact of the different samples was measured in order to calibrate the relationship between applied voltage and quantum dot carrier concentration. The capacitance of sample C is shown in Fig. 2 as a function of the applied bias. The resonances observed in capacitance reflect the successive loading of the quantum dots states with electrons.[12,8] Starting from the negative voltages, the first resonance at -0.78 V corresponds to the loading of one electron into the s ground state of the first quantum dot layer. The second capacitance resonance, at -0.63 V, corresponds to the loading of a second electron in the ground state of the dots. This peak is shifted in energy due to the Coulomb-blockade effect.[12] Because of the broadening of the peaks, the loading of the ground state of the second layer is masked by the capacitance associated with the first layer and cannot be clearly distinguished.[13] For these negative gate voltages, the electric field shifts the ground states of the quantum dots far from resonance with one another, and the electronic states are mostly localized in each quantum dot. Above -0.3 V, the loading of the *p*-excited states is observed. An average occupation of six electrons per dot in the first layer is obtained for a gate voltage around 0.2–0.3 V. Considering the lever arm[8] ([distance of the quantum dots above the backgate]/[depth of the backgate] $\sim 1/7$) and the interaction between both layers,[13] the loading of the electrons in the $s(p)$ states of the second layer is expected to occur around -0.27 V (0.27 V). A weak shoulder is effectively observed around 0.25 V on the capacitance spectrum. At higher voltages (0.5 V), the increase of the capacitance indicates the occupation of the two-dimensional states of the wetting layers. The flatband condition occurs for a gate voltage close to 0.7–0.8 V, when the gate voltage offsets the Schottky barrier. Note that in this case, the *p* states of both layers are occupied and can become hybridized between bonding and antibonding states.

Capacitance–voltage spectra for samples A and B are shown in the inset. The first electrons are loaded into the first dot layer in samples A and B at higher voltages (0.15 and -0.4 V, respectively) than in sample C, indicating that the ground states of the quantum dots are closer to the band edge of GaAs in samples A and B. This feature is attributed to a larger confinement energy which is expected from the reduced vertical thickness of the first layer of dots in sample A and to a slight variation of composition or height for sample B. The loading of the electrons in the *s* (*p*) states of the second layer of sample B is expected to occur around 0.01 V (0.58 V).

The normalized far-infrared transmission of sample B is shown in Fig. 3 for different gate voltages. The inset shows a comparison between the normalized transmission of sample B and sample A. The normalized transmission is defined as the ratio between the transmission at a given gate voltage divided by the transmission measured for a negative gate voltage (-1 V) where the quantum dots are empty. Two absorption resonances can be observed in Fig. 3. The first weak absorption resonance, with a maximum around 22 meV, is observed for gate voltages larger than 0.6 V. The second absorption resonance has a peak around 12 meV, becomes as large as 5.5%, and is clearly observed for gate voltages above 0.65 V. For gate voltages larger than 0.7 V, the spectra remain unchanged, thus indicating that higher biases do not affect the charging condition of the quantum dots.[14] The 22 meV absorption is also observed in sample A under similar bias conditions. However, as indicated by the inset, the broad absorption band maximum at 12 meV is clearly not present in sample A, thus ruling out the origin of the absorption as a defect absorption.

We have identified four processes which could possibly give rise to an absorption peak at THz frequencies in samples A and B: (1) transitions between the tunnel-split energy levels of the coupled wetting layers, (2) bound-to-continuum transitions between the *p* states of quantum dots and the wetting layers, (3) intersubband transitions in an accumulation layer which might be formed at high positive gate biases at the GaAs/AlAs interface just above the quantum dot layers (see Fig. 1), and (4) transitions between the tunnel-split states of the vertically coupled quantum dots. We can safely discard the $s-p$ transition as a possible source of absorption around 10 meV. Photoluminescence shows that the in-plane confinement is not significantly modified by the vertical

stacking of the quantum dots. The $s-p$ transition is therefore expected to occur around 50 meV.[8] Both samples A and B exhibit an absorption near 22 meV. A simple calculation (i.e., 0.5-nm-thick two-dimensional InAs layers separated by 7-nm-thick GaAs barrier) predicts the splitting of the bound states of the wetting layers to be at 24 meV. We therefore assign the 22 meV absorption to (1), transitions between tunnel-split states of the wetting layers, and turn our attention to the broad absorption near 12 meV observed in sample B but not in sample A. This is unlikely to be associated with (2), bound-to-continuum transitions between p states and wetting layers. In this case, the transition energy would shift significantly as more carriers are added in the quantum dots. Such transition would also be observed in sample A where, at flatband, electrons are loaded in the s and in the s and p states in the first and second layer, respectively. It is also unlikely that the 12 meV absorption is associated with (3), intersubband transitions in an accumulation layer.[15] Previous studies have not observed intersubband absorption from such an accumulation layer in samples containing quantum dots. If such an absorption were present in these samples, it would be expected in both samples A and B. The remaining possibility is (4), transitions between the tunnel-split ''bonding'' and ''antibonding'' states of the vertically coupled quantum dots. This assignment explains the following observations. The absorption is not observed in sample A since the quantum dot sizes are too dissimilar. This bonding-antibonding absorption can only be observed for voltages close to the flatband condition, since it requires the quantum dots to be loaded with electrons while the energy levels are brought into resonance and consequently split between bonding and antibonding states. The bonding-antibonding absorption is redshifted with respect to the wetting layer absorption since the tunnel-splitting depends on the confinement energy and decreases as the levels are more deeply localized in the wells. The distribution of dot sizes broadens the bonding-antibonding resonance to its width of 8 meV. Since the ground states of the quantum dots are expected to be filled with electrons at gate voltages higher than 0.6 V, the 12 meV intraband absorption is attributed to a transition between the split first excited states.

Figure 4 shows normalized transmission spectra in sample C, which are similar to those of sample B, thus supporting our assignments. A broad absorption band near 10 meV appears for a gate voltage of $V_g = 0.45$ V. The amplitude of the absorption is maximum for $V_g = 0.7$ V. Above this gate voltage, the 10 meV absorption is quenched progressively and finally completely disappears above 1.1 V. Another absorption above 20 meV grows with increasing gate bias. The absorption near 10 meV is again assigned to the transition between the tunnel-split p states of the coupled dots. This absorption is at a slightly lower energy than that observed in sample B, consistent with the barrier thickness being slightly larger (7.5 vs 7 nm), and the quantum dot states being somewhat deeper. A small energy shift of the transition as a function of the gate voltage is attributed to the potential drop between the coupled quantum dot layers. The absorption above 20 meV is assigned to the transition between tunnel-split states of wetting layers.

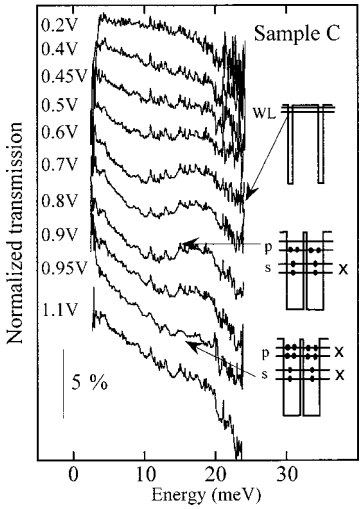

FIG. 4. Normalized far-infrared transmission of sample C as a function of the applied gate voltage. The curves have been offset for clarity. The inset explains schematically the spectra evolution.

The disappearance of the 10 meV absorption above 1.1 V in sample C suggests that the p states are completely filled at these gate voltages. This quenching is in agreement with capacitance spectra which indicate that all the p states (eight electrons) are likely to be populated in sample C. On the other hand, in the sample B dots with higher confinement, the charging effects prevent the loading of all p electrons and the THz absorption spectrum does not change above 0.85 V.

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