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Saturation of THz-frequency intraband absorption in InAs/GaAs quantum dot molecules

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We have investigated the far-infrared absorption in InAs/GaAs quantum dot molecules. The quantum dot molecules consist of two vertically coupled InAs self-assembled quantum dots separated by a GaAs barrier. The electronic coupling between the dot states results in an intraband absorption at THz frequencies. We show that this absorption can be bleached under high excitation intensity delivered by a free-electron laser. The saturation intensity is found to be on the order of 1 W cm^{-2}. The electron relaxation time T_1 is estimated from the saturation intensity. A lower limit for T_1 of the order of 30 ps is deduced. © *2000 American Institute of Physics.*
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The study of the electronic relaxation rate in quantum dots has attracted much interest in recent years. This interest was motivated by the prediction that the relaxation rate should be reduced in quantum dots because of the δ-like density of states of nanostructures.[1] The intradot relaxation rate depends on many parameters, such as the energy splitting between the confined levels and the carrier density. Several relaxation mechanisms, including phonon, multiphonon[2] or Auger-like relaxation[3] have been considered in the literature for carrier relaxation. Slowing of the intradot relaxation rate has been observed experimentally by time-resolved photoluminescence experiments in self-assembled quantum dots.[4] Note that a significant slowing of the relaxation rate has not been systematically observed. Several reports have been recently published on intradot relaxation time in the picosecond time scale, either by interband optical measurements[5] or by saturation of the intraband absorption in the midinfrared.[6]

It is well known that in semiconductor quantum wells, the electronic relaxation rate depends strongly on intersubband energy splitting. If the energy splitting is above the optical phonon energy, the relaxation typically occurs on a picosecond time scale through efficient optical phonon emission.[7] When the intersubband energy is below the optical phonon energy, the relaxation of cold carriers proceeds via acoustic phonon emission and the relaxation rate is strongly reduced. However, it has been shown that the intersubband relaxation time for such small intersubband splittings depends on the electronic temperature and on the energy distribution of the carriers.[8] Intersubband relaxation times ranging between tens of ps to ns have been reported depending on the experimental conditions.[9]

Intraband optical absorption at low energy (i.e., below the optical phonon energy) has been reported in semiconductor quantum dots obtained by standard lithography techniques.[10,11] Intraband absorption in the THz-frequency range can also be obtained by electronic coupling of self-assembled quantum dots.[12] Such vertically coupled quantum dots have been proposed as quantum bits for quantum computing.[13] In this letter, we show that the intraband absorption between electronically coupled quantum dots can be bleached under high pump excitation intensity.

The investigated quantum dot molecules consist of vertically coupled InAs self-assembled quantum dots grown by molecular beam epitaxy on a semi-insulating substrate.[14] The quantum dot layers of sample A (B) are separated by a 7 (7.5) nm thick GaAs barrier. The quantum dots are embedded in a field effect heterostructure. The structure consists of *n*-doped back contact, a 25 nm thick GaAs spacer layer, the coupled quantum dots, a 30 nm GaAs layer, a 108 nm thick AlAs/GaAs blocking barrier, and a 5 nm thick GaAs cap layer. The In(Ga)As quantum dots are lens shaped with a typical height of 6 nm and a diameter of 20 nm. An Al Schottky gate was deposited on top of the structure. This Schottky barrier allows the control of the electronic population of the quantum dots along with the electronic coupling between the quantum dot states. For far-infrared measurements, performed at liquid helium temperature, the samples were cleaved in 7 mm long bars and the infrared light was injected through the cleaved facet of the sample. The saturation experiments were performed with the far-infrared free-electron laser available in Santa Barbara.

Figure 1 shows the far-infrared transmission spectra measured by Fourier transform infrared spectroscopy for samples A and B. The transmission under a positive bias voltage was normalized by the transmission recorded at a negative bias voltage (-1.5 or -1 V) where the quantum dots are empty. The positive applied voltage allows one to populate the quantum dots and to achieve the flat band condition necessary to electronically couple the levels. A schematic description of the conduction band diagram is shown for a 0 V applied bias in the inset of Fig. 1. As reported in Ref. 12, the broad absorption which is peaked around 100 cm^{-1} is associated with the intraband absorption between the bonding and antibonding states of the coupled quantum dots.

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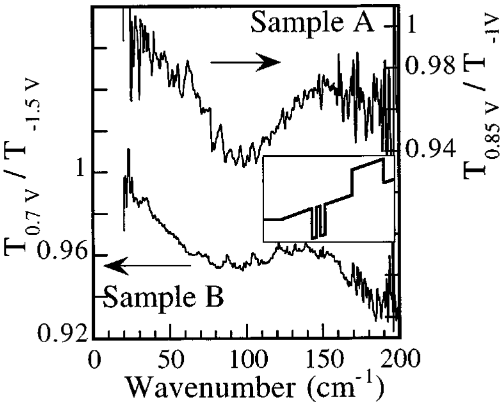

FIG. 1. Normalized transmission spectrum measured by Fourier transform infrared spectroscopy of samples A (top curve, 0.85 V gate bias) and B (bottom curve, 0.7 V gate bias). The inset shows a schematic of the band diagram under a 0 V bias condition.

As the *s* ground states of the dots are completely filled at a bias voltage around 0.8 V, the intraband absorption is attributed to transitions between the first excited bonding and antibonding *p* states. The quantum dot absorption is superimposed on top of a free-carrier absorption and an absorption between the coupled wetting layer states. At 85 cm^{-1}, the fraction of the absorption attributed to the quantum dots is $4 \pm 0.3\%$ ($2.2 \pm 0.3\%$) in samples A (B), as deduced by substracting the background absorption. The dipole matrix elements of the intraband absorption can be deduced from the integrated absorption and from the number of quantum dots as measured by capacitance spectroscopy.[12] A 12 nm (± 0.3) and a 5.5 nm (± 0.2) matrix element was deduced for samples A and B, respectively. The difference between these two numbers is likely attributed to the uncertainty of the optically active dot density. It is worth noting that the loading of the electrons does not occur at the same bias between both samples.[12] This feature indicates that a slight variation of composition or height exists between samples A and B. In the case of sample A, the amplitude of the intraband absorption between the bonding and antibonding states increases as the applied bias increases and saturates for bias values close to the flat band conditions. In the case of sample B, the amplitude of the intraband absorption is maximum at around 0.7 V and is quenched at higher bias when the quantum dots are fully occupied. In the following, we have performed the saturation measurements for an applied bias corresponding to a maximum of the intraband absorption (0.85 and 0.7 V for samples A and B, respectively). A reference measurement at a voltage (0.9 V) where the quantum dot absorption is quenched was also taken for sample B.

The amplitude of the normalized transmission at a fixed energy (85 cm^{-1}) is shown in Fig. 2 as a function of the applied gate bias. The full line corresponds to the absorption as measured by Fourier transform spectroscopy while the diamonds (squares) correspond to the normalized transmission measured with the free-electron laser at low excitation density for sample A (B). As seen, a satisfying agreement is obtained between both measurements. Though the absorption remains weak, it can be measured easily with the free-electron laser since the absorption is given by the transmission ratio at two different biases.[15]

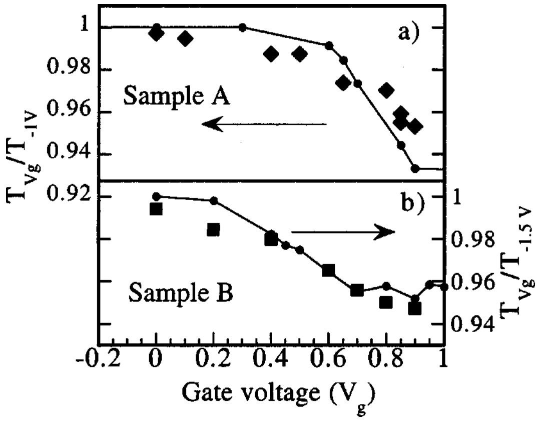

FIG. 2. Amplitude of the normalized transmission at 85 cm^{-1} of samples A and B as a function of the applied bias. The full lines correspond to Fourier transform absorption spectroscopy. The squares and the diamonds correspond to the normalized transmission measured with the free-electron laser.

Figure 3 shows the normalized transmission of sample A as a function of the free-electron laser intensity. The energy of the laser was set at 85 cm^{-1}. The transmission was measured for a 0.85 V applied bias. A fraction of the absorption clearly saturates at high intensity and is bleached above 100 W cm^{-2}. We attribute this bleaching to the saturation of the intraband absorption between the coupled quantum dot levels. The normalized transmission does not reach unity since the background absorption is not bleached for the investigated intensity range. The full curve in Fig. 3 is a least-squares fit using a standard absorption saturation law for an *inhomogeneously* broadened transition.[16] A saturation intensity of $0.25 \pm 0.15\,\mathrm{W/cm^{-2}}$ is deduced from the measurement. This saturation intensity is of the same order of magnitude as the saturation intensity reported for THz-frequency intersubband transitions.[17] It is worth noting that this intensity is 6 orders of magnitude lower than the one measured for intraband transitions in quantum dots at around 10 μm wavelength.[6] This feature is explained by the difference in the relaxation times and in the dipole matrix elements.

The normalized transmission of sample B is shown in Fig. 4 as a function of the pump excitation intensity. Figures 4(a) and 4(b) corresponds to a 0.7 and 0.9 V gate bias, re-

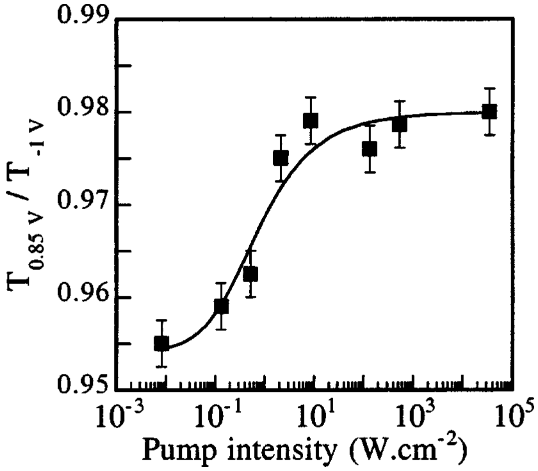

FIG. 3. Normalized transmission at 85 cm^{-1} of sample A vs the pump intensity. The gate bias is 0.85 V. The full curve is a fit which accounts for a standard absorption saturation law.

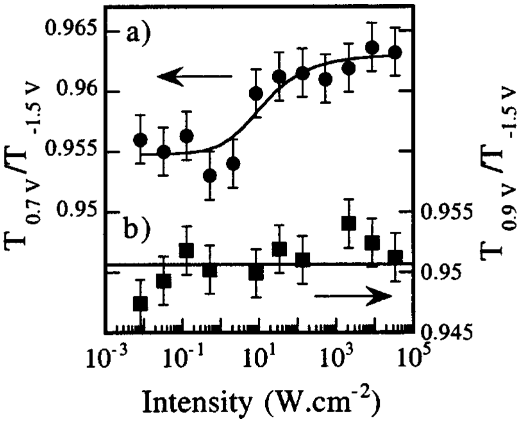

FIG. 4. Normalized transmission at 85 cm^{-1} of sample B vs the pump intensity. The gate biases are 0.7 V (a) and 0.9 V (b). The two vertical curves have been offset for clarity.

spectively. At 0.9 V, the quantum dot absorption of this sample vanishes due to the complete filling of the coupled states. Only the residual absorption (i.e., not quantum dot related) contributes to the transmission variation at 0.9 V.[18] As expected, effectively no dependence of the absorption versus the pump excitation intensity is observed at 0.9 V. Saturation of the absorption is, however, observed for a 0.7 V applied bias. This saturation corresponds to the bleaching of the intraband absorption of the coupled quantum dots. A saturation intensity of $4.5 \pm 3.5\,\mathrm{W\,cm^{-2}}$ is deduced from the measurement on sample B. We note that the magnitude of the bleaching for sample B is less than 1% while the quantum dot absorption is estimated to be more than 2% (see Fig. 1). This discrepancy may arise from a rectification effect which occurs at high pump intensities. The current through the structure increases with increasing free-electron laser intensity, indicating that the applied bias is effectively higher at these intensities. Since the absorption of the quantum dots decreases with increasing bias due to state filling,[12] the quantum dot absorption at the highest intensities is also decreased.

The saturation intensity measured for samples A and B is inversely proportional to the product of the relaxation (T_1) and dephasing (T_2) times. This product is found equal to 5500 and 1700 ps^2 for samples A and B, respectively. Part of the difference between these numbers is likely to be given by the uncertainty in the coupled far-infrared intensity. It is, however, difficult to extract the relaxation time unambiguously since the dephasing time is not experimentally known. A lower limit can nonetheless be estimated. The longest coherence time achievable is given by the dephasing associated with the energy relaxation. In the latter case, the coherence time T_2 is equal to $2T_1$. Assuming that this relation holds, the relaxation time T_1 is found equal to 50 (40–85) ps and to 30 (20–65) ps for samples A and B, respectively, if we account for the uncertainty on the saturation intensity deduced from the least-squares fit. An additional uncertainty of a factor of 2 also exists on the absolute value of the coupled intensity (30%–40% for T_1). It is likely that the dephasing rate of the optical coherence is faster than the recombination rate. The dephasing could be dominated by lattice relaxation mechanisms (i.e., coupling between electrons and acoustic phonons during the optical transition) which could occur on a shorter time scale.[19] Another source of broadening is related to the high number of carriers which are injected in the structure. The bonding–antibonding absorption can only be observed in flat band conditions. In this case, carriers are also injected into the two-dimensional wetting layer. The intraband transition could therefore be homogeneously broadened by the carrier–carrier interactions with the carriers in the two-dimensional gas contained in the wetting layer.[20] If the homogeneous broadening of the quantum dot transition is reduced, the relaxation time deduced with the saturation intensity would be consequently increased. The values of 50 and 30 ps obtained for samples A and B therefore represent a lower limit of the relaxation time.

The relaxation times deduced from the saturation intensity remain much shorter than those theoretically predicted by the interaction with acoustic phonons.[1] This feature can be explained by the high carrier density which is injected in the dots and in the wetting layer in order to observe the intraband absorption. This situation is the most favorable to enhance Auger-like scattering mechanisms. Very fast intradot relaxation times on the order of tens of ps have been theoretically predicted when a high two-dimensional carrier density is close to the quantum dots.[3,21]

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