Observation of Phonon Bottleneck in Quantum Dot Electronic Relaxation

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Time-resolved differential transmission measurements of self-assembled $In_{0.4}Ga_{0.6}As$ quantum dots clearly indicate a phonon bottleneck between the $n = 2$ and $n = 1$ electronic levels. The key to this observation is the generation of electrons in dots where there are no holes so that electron-hole scattering does not mask the bottleneck. We use a simple carrier capture model consisting of two capture configurations to explain the bottleneck signal and offer arguments to rule out other possible sources of the signal.

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It is well known that a "phonon bottleneck," or a suppression of carrier relaxation rates, is predicted for zero-dimensional semiconductor systems which have a discrete density of states [1,2]. If the energy level spacings are sufficiently large, then the rate of phonon-mediated scattering processes will be inhibited because energy and momentum conservation forbids transitions mediated by a single phonon emission. Semiconductor self-assembled quantum dots exhibit strong three-dimensional carrier confinement and show discrete energy spectra. Thus, it is predicted that relaxation rates among the electronic levels of these quantum dots would be low [3]. Since the time of the initial proposals, the search for the phonon bottleneck in self-assembled quantum dots has yielded no direct evidence supporting this prediction. Our initial measurements involving direct injection of carriers into the dot excited state showed a fast intersubband relaxation that is consistent with intradot electron-hole scattering [4]. Other groups investigating self-assembled quantum dots have also observed that the phonon bottleneck is circumvented, attributing this to a number of processes including Auger-type processes [5–7], intradot electron-hole scattering [3,8], and multiphonon processes [5,9]. The key to the observation of the bottleneck is then to remove all of these relaxation processes.

This can be accomplished by a carrier capture experiment in the following way. If electron-hole pairs are photoinjected into the continuum above the quantum dots, and the number of carriers is much lower than the number of accessible dots, the carrier capture process will occur mainly in two different configurations (see Fig. 1). One is a paired or geminate capture in which both the electron and the hole settle into the same dot. The other is an unpaired or nongeminate capture in which the electron and hole fall into two different, laterally separated dots. Geminately captured electrons will undergo fast relaxation due to electron-hole scattering, but electrons which are nongeminately captured into a quantum dot excited state will not experience electron-hole scattering and therefore will display a bottleneck in the relaxation. Previous experiments have not been sensitive to this process. We report here high-sensitivity nondegenerate pump-probe experiments which can observe both geminate and nongeminate populations using time-resolved differential transmission (DT) spectroscopy in $In_{0.4}Ga_{0.6}As$ self-assembled quantum dots.

The sample considered in this work is the same as the one used in our previous measurements [4,10] and similar to those used by us to realize state-of-the-art high-speed, low threshold quantum dot lasers [11]. It is an undoped heterostructure with four layers of $In_{0.4}Ga_{0.6}As$ quantum dots grown by molecular beam epitaxy. Each dot layer is separated by a 2.5-nm GaAs barrier layer. These four layers are sandwiched between two 0.1-μm thick GaAs layers and two outer 0.5-μm $Al_{0.3}Ga_{0.7}As$ carrier confinement layers. The entire structure is grown on a (001)

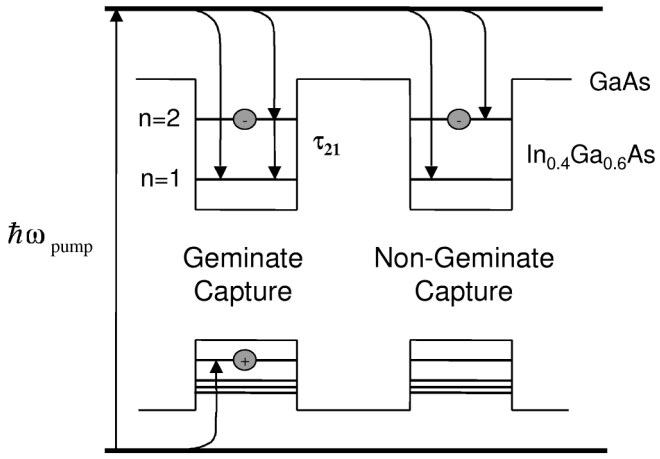

FIG. 1. Carrier capture model with geminate and nongeminate capture configurations.

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semi-insulating GaAs substrate which is subsequently removed through selective etching to enable DT measurements. The $In_{0.4}Ga_{0.6}As$ dots are grown at 520 °C while the rest of the sample is grown at 620 °C. Four dot layers are grown without generating additional defects to improve the dot uniformity and to increase the DT signal levels. High-resolution cross-sectional transmission electron microscopy (XTEM) shows near pyramidal dots with a base dimension of 14 nm and a height of 7 nm (see Fig. 2) [12], and cross-sectional scanning tunneling microscopy indicates carrier confinement within dots from similar samples grown here [13]. Atomic force microscopy scans reveal a dot density of about 5×10^{10} cm^{-2} per layer. Additional discussion of the characterization of similar dot samples is given in Refs. [11,12].

Band structure calculations of individual $In_{0.4}Ga_{0.6}As$ quantum dots based on an eight-band $k \cdot p$ formalism predict two confined electronic levels and several hole levels [14]. The interband optical transition probabilities are high only for transitions between electron and hole levels of the same quantum number. This selection rule was confirmed through spectral hole burning experiments extending the results in Ref. [10], which showed that a pump resonant with the excited states does not produce an instantaneous spectral hole involving the ground states. In real quantum dot ensembles, the discrete levels are inhomogeneously broadened due to the size variation of the dots. In multilayer structures, the quantum dots are vertically aligned, leading to vertical coupling between the electronic excited states [10]. Carrier-density dependent photoluminescence (PL) data on this sample support the existence of quantum confined dot states and confirm that the excited state interband transition (E2H2) is centered at 920 nm (1.35 eV) while the ground state transition (E1H1) is centered near 980 nm (1.27 eV) [4].

The DT measurements are carried out at 40 K with a pump-probe setup. For tunable pump and probe pulses, white light sources are generated using the output of a 100-fs 250-kHz amplified Ti:sapphire laser. A 10-nm bandpass filter is used to select the pump and probe pulses during DT time scans. For DT spectral scans, the probe pulse consists of the near-infrared band between 850 and 1050 nm selected with a long-pass filter. We use a prism pair to compensate for group velocity dispersion to limit the relative group delay to about 100 fs within that spectral range. The pump is tuned to generate carriers either in the GaAs barrier states for carrier capture experiments or resonantly in the excited states of the dots. We detect the probe DT signal with a lock-in amplifier referenced to the 2-kHz mechanically chopped pump. The DT signal is directly proportional to the transient carrier occupation of the probed levels [4]. Thus, monitoring the DT signal as a function of the delay between the pump and the probe allows us to see the time evolution of the dot level population.

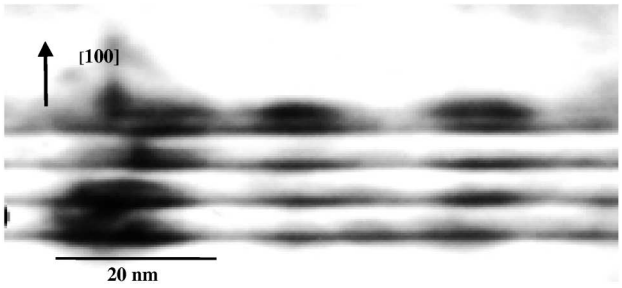

FIG. 2. Cross-sectional transmission electron microscope image of four-layer InGaAs quantum dots.

Results from our carrier capture experiments are shown on Figs. 3a and 3b. Here, the pump is centered at 800 nm and generates a carrier density that is less than one electron-hole pair per dot in the GaAs barrier layers. The subsequent carrier capture into and relaxation out of the excited ($n = 2$) state of the dot are monitored with a 910-nm probe. (A 910-nm probe is selected so that there are no DT contributions from the inhomogeneously broadened $n = 1$ transition, which shows no PL at 910 nm as reported in Ref. [4].) The $n = 2$ time scan shown in Fig. 3a reveals that, after a very fast capture into the dot excited state, some of the carriers relax quickly, as indicated by the fast decay component of the DT signal. But we see that the decay is not complete as is apparent from the long-lived signal, which persists beyond 40 ps. In longer time scans shown in Fig. 3b, we increase the delay time out to 600 ps and observe that this tail decays at a rate lower than the recombination rate (250 ps, as measured in a separate PL experiment). We believe that this slowly decaying signal is a clear sign of the predicted phonon bottleneck. If the phonon bottleneck

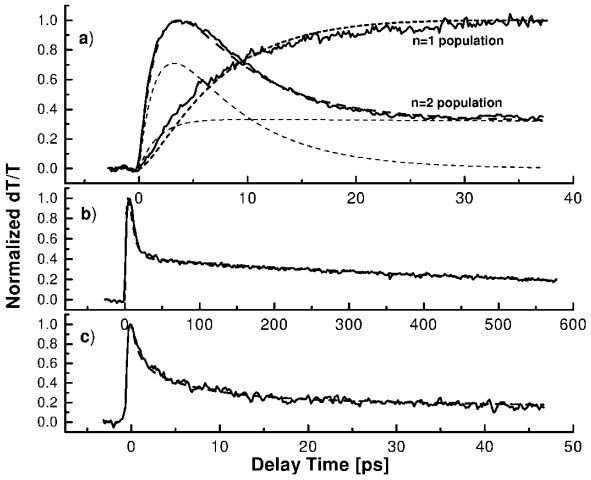

FIG. 3. DT time scans taken at 40 K. The rate equation fits are shown as dark dashed lines. (a) Nonresonantly pumped DT scan for $n = 1$ (980 nm) and $n = 2$ (910 nm) dot levels. The geminate and nongeminate components of the $n = 2$ fit are given as light dashed lines. (b) DT time scan of the $n = 2$ level with a long delay. (c) Resonantly pumped DT scan for the $n = 2$ dot level.

were nonexistent, one would expect the DT signal to decay quickly to zero as the excited level of the dot empties. Instead, we see a long-lived DT signal which reflects the extended occupation of the electronic excited state and, hence, the inhibition of relaxation processes.

As mentioned above, we understand this phonon bottleneck signal by attributing the origin of the fast and slow decay components to geminate and nongeminate capture processes, respectively. Within this model, one would expect the overall relaxation process to be composed of two parts. For the geminate capture case, intradot electron-hole scattering would dominate in producing a fast relaxation. In the nongeminate case, however, such scattering events would be suppressed because of the spatial separation of the electron and hole wave functions. Thus, the phonon bottleneck is observed as a result of this nongeminate capture which occurs when the carrier density is kept much less than one electron-hole pair per dot.

We next examine the phonon bottleneck at low carrier density [15] in time-resolved DT measurements with a resonant 920-nm pump at 40 K. In this experiment, since we place electron-hole pairs directly into the excited state of the dot, we expect the nongeminate configuration to be absent initially. The DT signal and a three exponential fit are given in Fig. 3c. After the excitation, the relaxation decay proceeds with an extremely fast time constant of 0.7 ps which we attribute to hole relaxation through phonon emission [8] and with another time constant of 6 ps which is consistent with an electron-hole scattering process as observed in our earlier measurements [4]. However, with our improved signal-to-noise ratio, we also see a definite long-lived signal as in the previous nonresonantly pumped case, even though the electrons and holes are injected geminately. Although the short length of our time scan allows for only a rough estimate, time constant values in the range of 100–200 ps fit the long-lived signal well and are comparable to recombination times [12] and to capture times in laser structures obtained from analysis of high-frequency electrical impedance data at room temperature [16]. This unexpected phonon bottleneck can result from electron-hole scattering which forms the nongeminate carrier configuration by kicking the initially excited electrons up into the wetting layer or the background continuum of states [17] and sending those electrons into dots without holes. If this lateral carrier coupling contributes to a nongeminate capture, the phonon bottleneck should be enhanced for carriers pumped higher in the dot well. This is what is indeed observed in measurements taken with a 910-nm pump that show a stronger bottleneck signal and corroborates our argument for a phonon bottleneck induced by lateral coupling. Thus, in both the resonantly and nonresonantly pumped cases, the phonon bottleneck is attributed to the nongeminate capture of carriers.

Applying the above carrier capture model and the intersubband relaxation time constant from the resonant pumping experiment, we fit the nonresonantly pumped data using two simple sets of rate equations. The first set accounts for the geminate capture configuration with a fast intersubband relaxation, while the second set describes the nongeminate capture case with a suppressed relaxation. The solutions to these two components are shown as light dashed lines under the $n = 2$ data in Fig. 3a. Since there is a single reservoir representing the barrier region, the solutions for the two sets of rate equations are summed with equal weight to produce a best fit for each of the excited state and ground state DT time scan data. The optimization of the least square fitting routine is guided by the simultaneous fit of both the $n = 1$ and $n = 2$ time scans. The $n = 2$ data can be fit very well with an adjusted intersubband relaxation time constant τ_{21} of 7.0 ps and a barrier to $n = 2$ capture times of 2.5 and 8.5 ps for the geminate and nongeminate capture cases, respectively (dark dashed line in Fig. 3a). The bottleneck signal originating from the nongeminate capture component decays with a time constant of 750 ps (Fig. 3b). Recombination terms from the barrier, $n = 2$, and $n = 1$ levels are also included in the rate equations although they do not play significant roles in the early transient dynamics. The best fit for the $n = 1$ signal absolutely requires direct (barrier to $n = 1$) capture terms with time constants of 30 ps for the geminate case and 100 ps for the nongeminate case. As is apparent in Fig. 3a, this simple model offers a very good fit to both the $n = 2$ and $n = 1$ time scans.

We eliminate the possibilities that the phonon bottleneck signal may be due to some anomalous source by considering the following. First, we eliminate the possibility that the long-lived signal is originating from defects or impurities in the GaAs and AlGaAs heterostructure surrounding the dots by performing the same carrier capture measurements on a control sample that is exactly the same except it contains no dots. In Fig. 4, we contrast the spectral scan from this dotless sample to that of our four-layer dot sample taken under the same conditions. As is evident, the dotless sample produces no signal at the dot transition energies even at a high carrier injection density equivalent to about 13 electron-hole pairs per dot, while the four-layer dot sample produces a strong signal. This confirms that the barrier and the cladding layers are not the source of the long decay signal.

We also consider the possibility of signal contributions from strain-induced interfacial defects surrounding the dots. First, it is highly unlikely that such states would be resonant with the quantum dot excited state. Second, if the long-lived state observed in DT experiments is due to such a defect level, then the defects must dominate the optical response of the structure, but this is incompatible with our resonant-pumping DT data [4], which shows that the decay of the $n = 2$ DT signal corresponds to the rise of the $n = 1$ population. We also know that the states responsible for the DT and PL signals must be confined in the quantum dots because the infrared transitions due to interlevel transitions are very strong in similar

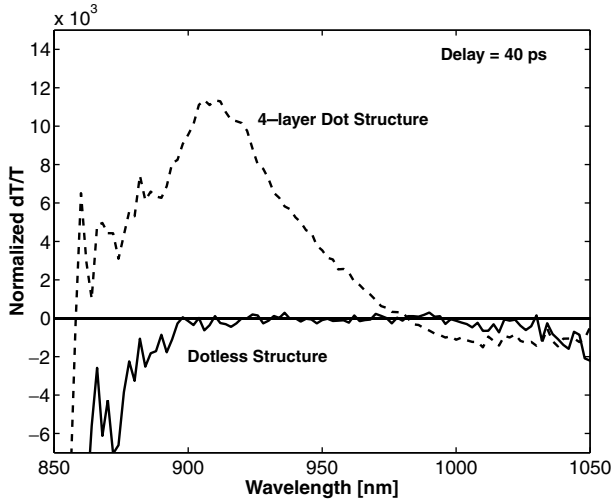

FIG. 4. Nonresonantly pumped DT spectral scans measured at 20 K for the dotless structure (solid line) and the four-layer dot structure (dashed line).

structures: high performance quantum dot intersubband lasers and detectors as well as interband lasers have been demonstrated [11,18], indicating that defects do not play a significant role in optical processes in these self-organized structures. Additionally, high-resolution XTEM and deep-level measurements do not show evidence of structural or other electrically active defects to be present.

Another possible source of the long-lived signal is thermal effects associated with lattice heating. When carriers are pumped into the GaAs barrier region with high excess energy, there is a possibility for lattice heating. The relatively slow energy dissipation for this heating may give rise to a slow DT signal. In our experiment, this possibility is very unlikely because of the low carrier densities encountered; the long-lived signal is observed for measurements in which the carrier density is as low as one electron-hole pair per ten dots. We confirm this by noting that, if the excess energy raises the lattice temperature and induces a change in the DT signal, that change in DT should scale with the lattice temperature. In contrast, in both resonantly and nonresonantly pumped DT time scans taken as a function of temperature, the amplitude of the long-lived tail remains approximately the same over the temperature range of 10 to 80 K. We verify that the excitation does not yield a lattice temperature beyond 80 K by calculating that the total carrier excess energy produces a temperature change of approximately 0.1 mK over the excitation volume. Thus, we rule out lattice heating as the source of our DT signals. (The temperature independence of the DT signal also indicates that hole thermalization does not make a contribution to the long-lived signal at lower temperatures.)

We note that another mechanism that can give rise to a long tail in the $n = 2$ signal could be the presence of an unintentional doping of the dots; due to Pauli blocking some of the carriers captured or injected into the $n = 2$ level would remain there instead of decaying down to the $n = 1$ level. We offer two arguments against this possibility in our experiment. First, the presence of a background carrier density would result in IR absorption. This has not been observed in undoped samples similar to ours. Second, if unintentional doping is playing a role in Pauli blocking, the ratio of the peak to the tail amplitude in the $n = 2$ DT signal would not change depending on whether the barrier or the $n = 2$ state is pumped. The fact that this ratio changes in our measurements is direct evidence for nongeminate capture.

We conclude from our time-resolved DT measurements that at low carrier densities and at low temperatures a phonon bottleneck originating in nongeminate capture is observed in $In_{0.4}Ga_{0.6}As$ quantum dots. This is the first time that such a predicted suppression of carrier relaxation has been observed in self-assembled quantum dots.

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