Temperature-dependent carrier dynamics in self-assembled InGaAs quantum dots

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We measured the transient temperature-dependent carrier population in the confined states of self-assembled $In_{0.4}Ga_{0.6}As$ quantum dots as well as those of the surrounding wetting layer and barrier region using differential transmission spectroscopy. Results show directly that thermal reemission and nonradiative recombination contribute significantly to the dynamics above 100 K. We offer results of an ensemble Monte Carlo simulation to explain the contribution of these thermally activated processes. [DOI: 10.1063/1.1462860]

Recent device performance and photoluminescence (PL) measurements indicate that temperature-dependent mechanisms greatly affect carrier dynamics in self-assembled quantum dots.[1–5] Some of these reports claim that the temperature-sensitive operation of devices originates from nonradiative recombination (NR) and thermal reemission of carriers from the dot levels to the wetting layer (WL) and the barrier region.[6,7] This carrier reemission is believed to occur through thermal absorption and is strong at high temperatures because carriers couple to a large density of states in the WL.[8] Further measurements and a clear understanding of these temperature-dependent carrier dynamics in quantum dots are important.

We report here direct evidence of this thermally driven carrier dynamics measured through differential transmission (DT) pump–probe experiments. Specifically, we time-resolve the dot level population as well as that of the WL/GaAs barrier region to show directly that thermalization of the carriers occurs with significant reemission of carriers from the dot to the WL and the barrier states with increased temperature. In order to see these effects without the influence of complex Auger-type processes, the DT measurements are done in the low-carrier-density regime. Taking into account the temperature-dependent capture and relaxation rates, we perform an ensemble Monte Carlo analysis of the carrier dynamics to understand the observed temperature-dependent dynamics.

The sample for these experiments is an undoped heterostructure with four layers of $In_{0.4}Ga_{0.6}As$ quantum dots grown by molecular beam epitaxy with an areal dot density of about 5×10^{10} cm^{-2} per layer. The 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, and each layer is separated by a 2.5-nm GaAs barrier layer. Cross-sectional transmission electron microscopy shows near pyramidal dots with a base dimension of 14 nm and a height of 7 nm. Carrier-density dependent PL data on this sample support the existence of quantum confined dot states and show that the excited state interband transition ($n=2$) is centered at 920 nm (1.35 eV), while the ground state transition ($n=1$) is centered near 980 nm (1.27 eV).[9] PL data and calculations based on eight-band $k\cdot p$ formalism indicate that the electronic $n=2$ level is about 50 meV below the GaAs conduction bandedge. Additional discussions of the characterization of similar dot samples are given in Refs. 6 and 10.

The DT measurements are performed over a range of 10 to 290 K with a pump–probe setup. A 100-fs 250-kHz amplified Ti:Sapphire laser is used to generate white-light sources from which 10-nm wide pulses are spectrally selected for the pump and probe pulses. The pump is tuned to generate carriers either in the GaAs barrier states or resonantly in the excited states of the dots. For DT spectral scans, the probe pulse consists of a dispersion-compensated near-infrared band between 820 and 1050 nm selected with a long-pass filter. Using a monochromator, we resolve the DT signal to 1 nm and detect the probe signal with a lock-in amplifier referenced to the 2-kHz mechanically chopped pump. The DT signal measures the change in the carrier occupation of the levels that are in resonance with the probe spectrum. When the pump and probe pulses are delayed with respect to each other, the transient dot level population is resolved directly.[9] The temperature of the sample is stabilized in a He flow cryostat with a feedback-heater controller. When the temperature is varied, markers on the sample are used to maintain the same location, and the detection wavelength is redshifted to match the temperature-dependent spectral shift of the dot PL and DT spectra. In our sample, this shift of the confined dot levels amounts to only about 20 meV and the DT time scans do not show significantly different behavior between the shifted and nonshifted probes, especially at higher temperatures. This latter point may be due to the overall thermalization of the carriers among the levels.

In Fig. 1(a), we plot the $n=2$ (910 nm) DT time scans measured at 40–150 K. The pump pulse, centered at 800 nm, generates a carrier density that is less than one electron–hole pair per dot in the GaAs barrier layers. We have shown pre-

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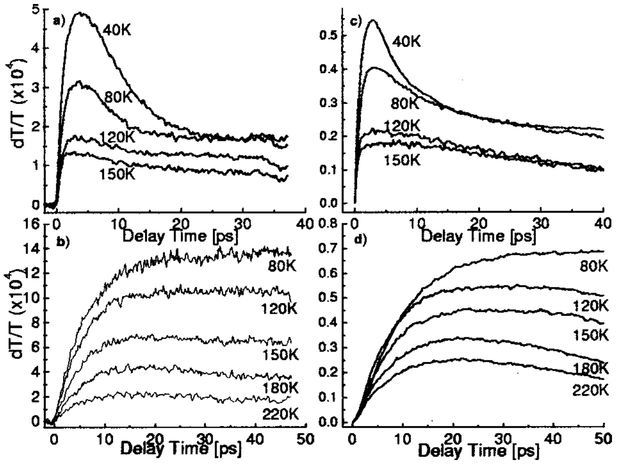

FIG. 1. Temperature-dependent DT time scans. (a) $n=2$ DT time scans taken at 40, 80, 120, and 150 K. (b) $n=1$ DT time scans taken at 80, 120, 150, 180, and 220 K. Monte Carlo simulation of (c) $n=2$ and (d) $n=1$ level population.

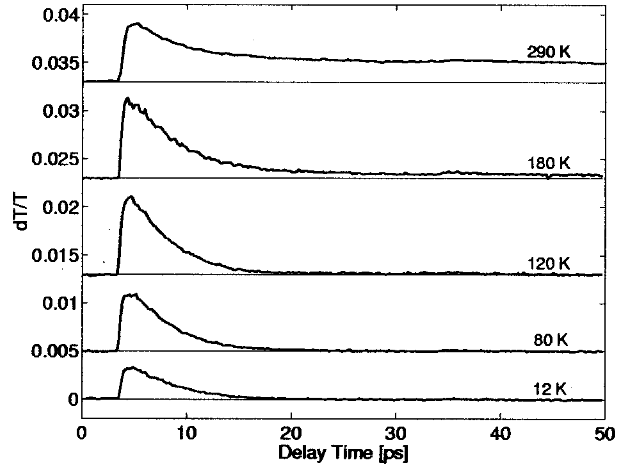

FIG. 2. DT time scan of wetting layer and barrier region (± 20 meV of GaAs bandedge) measured at 12, 80, 120, 180, and 290 K.

viously in Ref. 11 that in the low density regime, electrons and holes may be captured either geminately into the same dot or nongeminately into different dots. The rise time corresponds to the carrier capture into the excited state, and the initial fast decay in the low-temperature data arises from fast electron–hole scattering of geminately captured pairs. The long (>100 ps) component is from nongeminately captured electrons trapped in the excited state by the phonon bottleneck.[11] As the temperature is increased, the fast component of the $n=2$ DT signal decreases significantly in magnitude suggesting electron–hole scattering may play a role in the reemission; at 120 K, most of the geminate population disappears. As the temperature is increased beyond 120 K, the overall $n=2$ DT signal diminishes. This trend continues through room temperature (not shown here). The data for the $n=1$ (980 nm) DT time scans are given in Fig. 1(b) for the temperature range 80 to 220 K (below 80 K, the scans are very close to the 80 K DT scan in magnitude and shape). Here, we see the build up of the ground state population, which again decreases in magnitude at higher temperatures indicating signs of carrier reemission.

In order to observe directly the thermal reemission of the carriers from the confined dot states to the WL and barrier regions, we perform the DT scans on the energy states within ± 20 meV of the temperature-dependent GaAs bandedge. These results, shown in Fig. 2, indicate that at low temperatures, the carriers generated in the barrier region all relax into the low-lying WL and dot levels. Around 120 K, the carrier population begins to occupy the WL/GaAs barrier states, and above 180 K, a significant number of the carriers occupy these high-lying states. From spectral scans taken around the bandedge, we see that most of the carriers observed within the ± 20-meV detection window populate the barrier states. We also know that Pauli blocking is not occurring in the dot levels to produce this barrier signal because the spectral scans show that the overall DT spectrum of the dot levels decreases in magnitude at higher temperatures.

We model the temperature-dependent carrier dynamics observed using a detailed ensemble Monte Carlo simulation that accounts for random scattering events (for details see Refs. 12 and 13). The model consists of an ensemble of identical dots, each of which has an electron and hole energy spectrum obtained from a $k \cdot p$ band structure calculation.[14] The carrier capture, relaxation, and reexcitation occur among the two-dimensional GaAs barrier region, a linearized density of states of the WL, and the dot states. To obtain the results, we use the time constants of the individual decays as adjustable parameters. In an effort to simplify the model, we adjust only those parameters which capture the main features observed in the DT time scans. In Figs. 1(c) and 1(d), we provide a set of simulation results that well match the main dynamical features of the $n=2$ and $n=1$ DT data. For these results, we use an ensemble of 5000 dots and inject a carrier density of 0.5 electron–hole pairs per dot into the GaAs barrier. For increasing temperature, we reduce the capture times for the $n=2$ and $n=1$ electron (hole) from about 2.5 and 40 ps (50 ps) at low temperature to 2.0 and 20 ps (15 ps) at 290 K to account for the faster rise of the $n=2$ and $n=1$ populations. It is possible that this increased capture rate at high temperature is due to decreased diffusion time of carriers through the barrier region and the increased resonant capture of thermalized carriers from the WL/barrier region through multiphonon scattering.[15] The temperature-dependent electronic intersubband relaxation time constants were obtained directly from resonant-excitation DT measurements which remove the complications associated with the capture process. These resonant measurements indicate that the intersubband relaxation stays relatively constant at low temperature at 5–10 ps. For higher temperatures, it is difficult to extract the intersubband relaxation from the resonant data because reemission and NR greatly affect the decay. In the model, we keep this relaxation time constant fixed for simplicity.

As the temperature is increased, reemission and NR play a stronger role. In the 80-K $n=2$ time scan, the geminate component of the carriers undergoes reemission at a faster rate than the nongeminate population. This can be attributed to electron–hole scattering in which $n=2$ electrons are reemitted into the WL by acquiring energy from thermalizing holes. For the nongeminate electrons, the reemission is mediated by interactions with the lattice. Up to 80 K, the magnitude of the $n=1$ population remains relatively constant because the electronic intersubband energy spacing ($\Delta E_{21} \approx 60$ meV) is too large for reemission to occur from the ground state and the holes do not change their thermal distribution significantly. When the temperature is increased beyond 80 K, the $n=1$ DT signal decreases initially because the holes redistribute themselves due to thermalization. Then carriers are reemitted into the high-lying WL and barrier states causing a reduction in the dot DT signal. In adjusting the rate of reemission in the model, we monitor the resulting barrier population as a function of temperature; the reemission time constant from the electronic $n=2$ level ranges from hundreds of picoseconds at low temperature down to tens of picoseconds at 290 K. When this is done, we find that the level of signal decay in the $n=2$ and $n=1$ levels to be insufficient. Here, we invoke NR which accounts for carrier losses as observed in luminescence experiments. The nature and process for NR are very complex, and we consider simple exponential decays from the entire population while taking into account that the higher-lying states appear to be less affected by NR than the low-lying states as observed in the DT time scans. This produces the overall decrease in the DT signal for both the $n=2$ and $n=1$ levels, and accounts for the early decay of the $n=1$ population at high temperatures.

The DT time scans and our Monte Carlo model results show reasonable agreement. We emphasize the need for reemission and NR terms to account for the population of the barrier states and the decrease in the DT signal at temperatures above 100 K. The fact that the degradation of the threshold current and modulation rates of InGaAs quantum dot lasers[6] coincide with the onset of strong thermal reemission near 100 K leads us to believe that the thermally activated dynamics directly cause the degradation in laser performance. These issues will also need to be addressed for the high performance of near-, mid-, and far-infrared photodetectors that require low dark currents at room temperature.

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