Gain dynamics and ultrafast spectral hole burning in In(Ga)As self-organized quantum dots

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Using a femtosecond three-pulse pump-probe technique, we investigated spectral hole-burning and gain recovery dynamics in self-organized In(Ga)As quantum dots. The spectral hole dynamics are qualitatively different from those observed in quantum wells, and allow us to distinguish unambiguously the gain recovery due to intradot relaxation and that due to carrier capture. The gain recovery due to carrier–carrier scattering-dominated intradot relaxation is very fast ($\sim$130 fs), indicating that this is not the factor limiting the bandwidth of directly modulated quantum dot lasers.
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Self-organized quantum-dot (QD) active regions are being researched intensively for laser and amplifier devices because of their low threshold current density,[1] reduced temperature sensitivity,[2] and high differential gain.[3] Their potential for high-speed devices is also being studied: small-signal modulation bandwidths of ridge waveguide lasers of 5–7.5 GHz at 300 K and greater than 20 GHz at 80 K have been achieved.[4] Carrier dynamics in the QDs are critical to the understanding of these devices. We have previously used femtosecond time-resolved spectroscopy to study the carrier capture and intradot carrier relaxation in the low-density (absorption) regime, and have observed the effects of the phonon bottleneck[5] and intradot electron–hole scattering.[6] In the gain regime at high carrier density, however, carrier–carrier scattering processes become more dominant; fast gain recovery ($\sim$100 fs) dynamics have been previously observed in an InAs–InGaAs QD active waveguide, using a $p-i-n$ ridge structure at room temperature.[7] In the experiments reported here, we use femtosecond white light spectroscopy to study spectral hole burning in both the excited and ground states, which directly shows the carrier relaxation between the discrete energy levels in the QDs. The gain recovery due both to carrier relaxation from the confined $n=2$ state and to carrier capture from the barrier region are observable in our spectrally and temporally resolved experiments.

Two samples are used in this work: $In_{0.4}Ga_{0.6}As$ and InAs QDs. The $In_{0.4}Ga_{0.6}As$ (InAs) sample is an undoped heterostructure with four (five) layers of $In_{0.4}Ga_{0.6}As$ (InAs) quantum dots, separated by 2.5 nm (2 nm) GaAs barriers, grown by molecular-beam epitaxy. These 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 structures are grown on (001) semi-insulating GaAs substrates which are subsequently removed through selective etching to enable differential transmission (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. Cross-sectional transmission electron microscopy shows that the dots are pyramidal in shape with a base dimension of 14 nm and a height of 7 nm. Atomic-force microscopy scans reveal a dot density of 5×10^{10} cm^{-2} per layer. For all the data reported here, the sample is held at a temperature in the range of 8–15 K, in order to eliminate the effects of thermal excitation and lateral interdot coupling which occur at higher temperature. The ultrafast gain dynamics at room temperature occur on a slightly faster time scale, and will be reported elsewhere.

Band-structure calculations of individual QDs based on an eight-band $\mathbf{k}\cdot\mathbf{p}$ formalism predict two strongly confined electronic levels and a larger number of hole levels.[8] The interband transition probabilities are high only for those transitions between electron and hole levels of the same quantum number. In real QD ensembles, these discrete levels are inhomogeneously broadened due to the size fluctuation of the dots. The excited level in each dot has a two-fold degeneracy due to the symmetry of the dot geometry, in addition to the double-spin degeneracy. At a low temperature, the excited state interband transition $/E2H2/$ is centered around 930 nm (980 nm)[9] in the $In_{0.4}Ga_{0.6}As$ (InAs) sample, and the ground state transition $/E1H1/$ is centered at 975 nm (1000 nm) in the $In_{0.4}Ga_{0.6}As$ (InAs) sample. Figure 1(a) shows DT/T spectra (i.e., DT spectra normalized to the transmitted probe spectrum) of the $In_{0.4}Ga_{0.6}As$ sample measured using a 100 fs white-light probe pulse following excitation by an optical (800 nm) pump pulse for different pump fluences; saturation of the carrier population in the ground state is clearly observed.

Femtosecond three-pulse white-light pump and probe DT spectroscopy is performed using an 85 fs, 3.5 μJ, 250 kHz Ti:Sapphire regenerative amplifier system. Carriers are injected by optically pumping the GaAs barrier region (800 nm, ''gain pulse'') to establish a population inversion in QDs; the intensity of the gain pulse is adjusted so that the dots can be in the absorption or gain regime. Tunable pump

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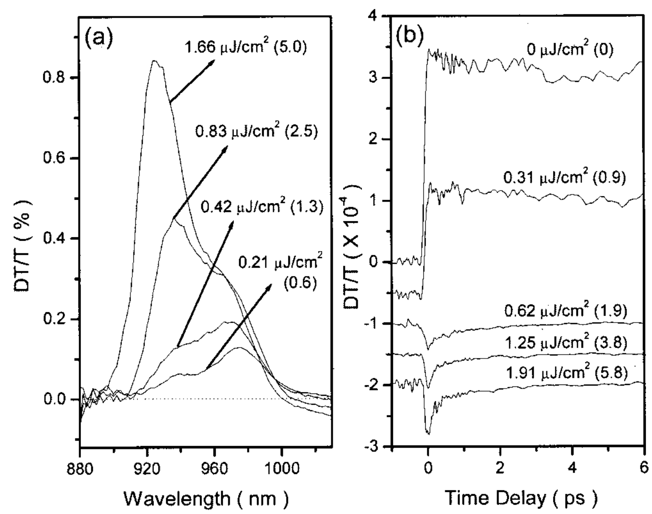

FIG. 1. (a) DT/T spectra measured with a broadband white-light probe pulse following injection of carriers into the barrier regions by an 800 nm ''gain'' pulse at a probe delay of 14 ps for different gain pulse fluences, and (b) DT time scans measured with 970 nm pump and probe for different fluences of the gain pulse which injects the carriers at $t=-14$ ps. We give in parentheses the numbers of e–h pairs per QD injected into the $In_{0.4}Ga_{0.6}As$ sample by the gain pulse.

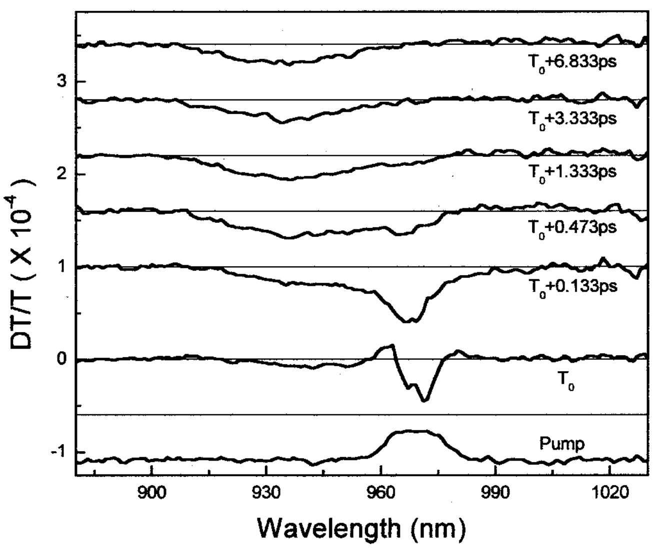

FIG. 2. DT/T spectra measured with a 970 nm pump and a broadband white-light probe in $In_{0.4}Ga_{0.6}As$ QDs versus probe delay (pump pulse at approximately $t=T_0$, gain pulse at $t=T_0-14$ ps).

and probe pulses are generated by spectrally filtering two single-filament white-light sources; a 10 nm bandwidth ''pump pulse'' is tuned to resonantly deplete (or generate) electron–hole pairs in the ground state by stimulated emission (or absorption) in the gain (or absorption) regime, after a 14 ps delay with respect to the gain pulse. The depletion (generation) of carriers by the pump pulse in ground state gives rise to a negative (positive) DT signal; this sign flip of the DT signal is the critical indication of gain in the QDs. The pump pulse is fixed at a 14 ps time delay with respect to the gain pulse, because the carrier population in the ground state is saturated at this time.

A broadband white-light probe measures the DT spectrum as a function of pump–probe delay. We select the spectral band between 880 nm and 1030 nm with an RG850 Schott filter, and use a prism pair to compensate for group velocity dispersion to limit the relative group delay to about 50 fs within this spectral range. Spectral components shorter than 850 nm in the probe are removed by a mask in the prism-pair arm to prevent any carrier generation in the GaAs barriers by the probe pulse. The pump pulse is mechanically chopped at 6 kHz, and the DT signal is measured using a lock-in amplifier; the probe DT is plotted as a function of delay following the pump pulse. The three pulses are focused on the QD sample near normal incidence; in this geometry, we obtain the intrinsic single-pass gain dynamics in the QDs, uncomplicated by any propagation effects or device parasitics that one might have in $p-i-n$ waveguide structure.

Figure 1(b) shows time-resolved degenerate pump–probe scans of the ground state ($E1H1$) DT signal in $In_{0.4}Ga_{0.6}As$ QDs for different gain-pulse fluence densities. The DT signal is positive (induced transmission) when the gain pulse fluence is zero, and becomes negative when the gain pulse exceeds the transparency fluence of 0.36 $\mu J/cm^2$ [$\sim$1.1 electron–hole (e–h) pair per dot]; this number is consistent with the data of Fig. 1(a), which shows that about half of the $n=1$ states are full at this excitation density. In the gain regime, the DT data show a fast recovery of the gain; the temporal behavior of the gain recovery is observed to be weakly dependent on the carrier density. The fast components for the gain fluences of 0.62, 1.25, and 1.91 $\mu J/cm^2$ are (170$\pm$19), (160$\pm$18), and (140$\pm$31) fs, respectively. The amplitudes of both the initial pump-induced gain depletion and the fast component of the gain recovery are larger for higher carrier densities. The numbers of e–h pairs per dot in parentheses in Fig. 1 are obtained from the measured spot size and power in the gain pulse using published values of the absorption coefficient ($\alpha\sim12\,045$ cm^{-1}) and reflectivity ($R\sim0.3285$).[10]

By spectrally resolving the probe, we obtained DT/T spectra in the gain regime at different probe delay times; results from $In_{0.4}Ga_{0.6}As$ QDs with a gain pulse fluence of 1.27 $\mu J/cm^2$ are shown in Fig. 2. Near time zero, the pump pulse burns a spectral hole in the gain spectrum centered at the pump wavelength by depleting the carrier population in the $n=1$ QD level. After about 200 fs, a satellite spectral hole appears at the $n=2$ excited state transition, which results when $n=2$ carriers relax to the $n=1$ state following the gain depletion induced by the pump pulse. These results support models which have proposed that QD excited state carriers are the reservoir for the optically active ground-state carriers resulting in subpicosecond gain recovery as long as the excited state is well populated.[11] It should be noted that the nature of the DT/T spectra in the gain regime in self-organized QDs and in quantum wells (QWs)[12] are qualitatively different; the spectral hole localized on the excited state is due to the discrete nature of the states in self-organized QDs. Most significantly, by observing the temporal evolution of the $n=1$ and $n=2$ spectral holes, it is possible to unambiguously distinguish between gain recovery due to relaxation between the discrete QD states and recovery due to capture from the barrier regions around the QDs.

The ground-state gain recovery dynamics of the $In_{0.4}Ga_{0.6}As$ QDs [solid line in Fig. 3(a)] exhibit a fast component (0.13 ps) and a slower component (1 ps). Similar time constants are observed for InAs QDs, although the precise

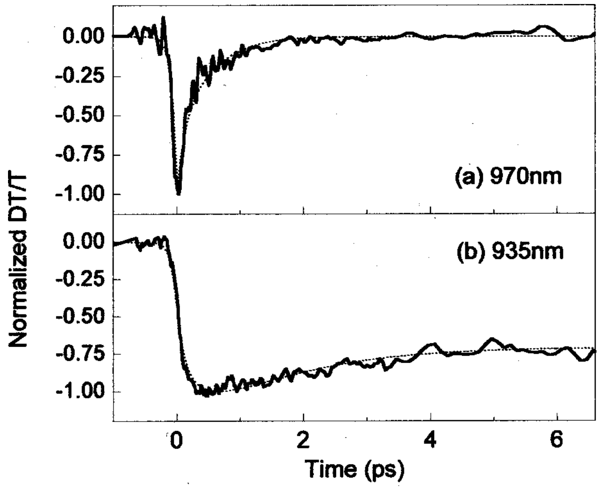

FIG. 3. DT signals in $In_{0.4}Ga_{0.6}As$ QD's measured with a 970 nm pump with the probe tuned to (a) 970 nm (QD ground state) and (b) 935 nm (QD excited state). The 800 nm gain pulse was 14 ps before the pump. The solid lines are the experimental data and the dotted lines are populations calculated with a rate equation model.

relaxation rates differ slightly (0.18 and 1.8 ps). Figure 3(b) corresponds to the carrier population in the excited state. The dotted lines in Figs. 3(a) and 3(b) are the carrier populations in the ground and excited states calculated using a three-level rate equation model (barrier plus $n=1$ and 2 confined states). The carrier recombination and thermal excitation to excited states are ignored as they are much longer than the gain recovery dynamics at a low temperature. The rate equation model does not distinguish electron and hole scattering separately (i.e., a single rate constant is assumed for the $n=1$ and $n=2$ gain recovery dynamics). The best fit is obtained using an $n=2$ state to $n=1$ state relaxation time (τ_{21}) of 130 fs and a carrier capture time (τ_{b2}) from the barrier to the $n=2$ state of 1 ps. The capture time is consistent with results we have obtained previously in low-carrier-density experiments,[6] indicating the capture is dominated by phonon-mediated relaxation. In contrast, τ_{21} in the gain regime is significantly faster than that observed when the population is much less than one pair per dot (5.2 ps for electrons and 0.6 ps for holes);[6] this indicates that carrier–carrier scattering is the mechanism responsible for intradot relaxation.

In conclusion, we have measured the gain recovery time constants in the ground state of undoped In(Ga)As QDs using a three-pulse pump–probe experiment. The spectral hole-burning data indicate that the subpicosecond gain recovery is due to intradot relaxation (~130 fs) via carrier–carrier scattering and the few-picosecond recovery component is due to phonon-mediated capture. These results imply that the intrinsic carrier capture and intradot relaxation are not the current limiting factor for high-speed performance. We have observed a localized spectral hole burning in the excited state which is not observed in QWs; this is the result of the zero-dimensional nature of the QD confined states.

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