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 (~130 fs), indicating that this is not the factor limiting the bandwidth of directly modulated quantum dot lasers. © 2002 American Institute of Physics. [DOI: 10.1063/1.1493665]

Self-organized quantum-dot (QD) active regions are being researched intensively for laser and amplifier devices because of their low threshold current density, reduced temperature sensitivity, and high differential gain. Their potential for high-speed devices is also being studied: small-signal modulation bandwidths of ridge waveguide lasers of 5–7 GHz at 300 K and greater than 20 GHz at 80 K have been achieved. 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 and intradot electron–hole scattering. In the gain regime at high carrier density, however, carrier–carrier scattering processes become more dominant; fast gain recovery (~100 fs) dynamics have been previously observed in an InAs–InGaAs QD active waveguide, using a p-i-n ridge structure at room temperature. 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: In0.4Ga0.6As and InAs QDs. The In0.4Ga0.6As (InAs) sample is an undoped heterostructure with four (five) layers of In0.4Ga0.6As (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 Al0.3Ga0.7As barrier 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 In0.4Ga0.6As 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 × 1010 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 k·p formalism predict two strongly confined electronic levels and a larger number of hole levels. 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 /E1H/ is centered around 930 nm (980 nm) in the In0.4Ga0.6As (InAs) sample, and the ground state transition /E1H/ is centered at 975 nm (1000 nm) in the In0.4Ga0.6As (InAs) sample. Figure 1(a) shows DT/T spectra (i.e., DT spectra normalized to the transmitted probe spectrum) of the In0.4Ga0.6As 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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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 RGG850 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 lock-in amplifier; the probe DT is plotted as a function of pump–probe delay. We select the spectrum as a function of pump–probe delay. We select the spectrum as a function of pump–probe delay. We select the spectrum as a function of pump–probe delay.

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 $n=1$ 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
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 (τ21) of 130 fs and a carrier capture time (τ21) 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, indicating the capture is dominated by phonon-mediated relaxation. In contrast, τ21 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), which 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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9. At very low density, the n = 2 absorption peak appears near 920 nm. When >1 e−h pair per dot is injected, there is a redshift in the n = 2 DT peak due to Coulomb many-body interactions.