Evidence of interdot electronic tunneling in vertically coupled $In_{0.4}Ga_{0.6}As$ self-organized quantum dots

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Ultrafast differential transmission spectroscopy with a resonant pump reveals evidence of electronic tunneling among the excited levels of vertically aligned $In_{0.4}Ga_{0.6}As$ self-organized quantum dots. This evidence of tunneling is observed as a rapid spectral redistribution of electrons within a few hundred femtoseconds of optical excitation. Measurements show that this spectral spread is independent of carrier density and, therefore, indicate that carrier–carrier scattering is not the main mechanism for carrier redistribution. Instead, electronic tunneling is responsible for the interdot coupling; tunneling rate calculations agree reasonably with the experiment, supporting this conclusion. © *2000 American Institute of Physics.* [S0003-6951(00)04417-X]

Time-resolved studies of ultrafast carrier dynamics in epitaxially grown self-organized quantum dots have contributed significantly to the understanding of the physical mechanisms that govern quantum-dot-based devices. For example, recent time-resolved optical studies show that the predicted phonon bottleneck is circumvented through Auger-like scattering processes[1–4] and as a result does not limit laser operation.[5] Some of these studies, including our previous work, look at quantum-dot systems as an inhomogeneous distribution of ideal, isolated dots which have discrete, uncoupled levels.[6] Following optical excitation, the initial transmission spectrum of such a system should show a spectral hole burned at the dot transition energies that are in resonance with the pump spectrum. As time evolves, the carriers relax from these high-energy states, and should form a replica spectral hole around the ground state. In self-organized quantum-dot devices, in order to enhance the optical gain in quantum-dot lasers or the responsivity of detectors, the dot areal density is maintained as high as possible, and multilayer structures are used. In multilayer structures with thin barrier regions, it is well known that the quantum dots are aligned vertically.[7] Therefore, one might expect coherent coupling of the electronic levels to occur,[8–10] which will significantly affect the initial evolution of the carrier distribution following optical excitation. The strong coupling between the dots has important implications for devices which are designed to exploit the quantum-dot δ-function-like density of states.

We report on our temporally and spectrally resolved differential transmission (DT) spectroscopy measurements of $In_{0.4}Ga_{0.6}As$ quantum dots, which show evidence of interdot coupling. In contrast to our previous work,[6] we examine the entire DT spectrum of the quantum dots simultaneously, which allows us to directly resolve the population dynamics within the inhomogeneously broadened band of levels. The carrier density dependence of the coupling dynamics is also tested to examine possible relaxation mechanisms. Our results indicate that the interdot coupling is due to tunneling of electrons among the excited states of vertically aligned quantum dots. Calculations of electronic tunneling rates between vertically coupled dots show reasonable agreement with our experimental results.

The sample considered in this work is an undoped heterostructure with four layers of $In_{0.4}Ga_{0.6}As$ quantum dots, separated by 2.5 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.[6] The entire structure is grown on a (001) 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. 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. In this arrangement, two vertically aligned quantum dots from adjacent layers are spatially closer to each other on average than two nearest-neighbor quantum dots in a single layer.

Previous band-structure calculations of individual quantum dots based on an eight-band $k\cdot p$ formalism predict two confined electronic levels and several hole levels.[6] The interband transition probabilities are high only for those transitions between electron and hole levels of the same quantum number. In real quantum-dot ensembles, these discrete levels are inhomogeneously broadened due to the size variation of the dots. In addition, level splittings occur due to interdot coupling, causing the formation of bands of electronic levels around the central excited- and ground-state levels. The excited level in each dot has a twofold degeneracy due to the symmetry of the dot geometry. In the four vertically coupled dot configuration, the excited levels form a band of eight levels each of which has a spin degeneracy of two. The

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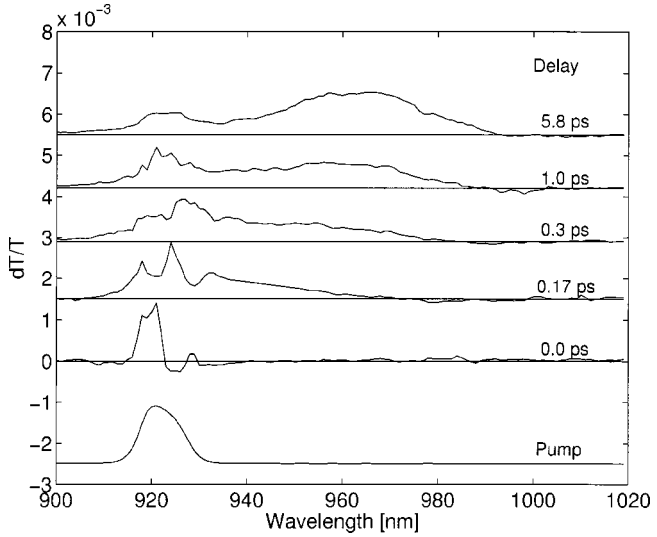

FIG. 1. Differential transmission spectra at delays of 0.0, 0.17, 0.3, 1.0, and 5.8 ps (bottom to top). The pump spectrum is given on the bottom of the figure. The rapid energy redistribution is evident in the 0.17 ps delay spectrum. The oscillations around the pump wavelength are due to the interference between the pump and the probe pulses.

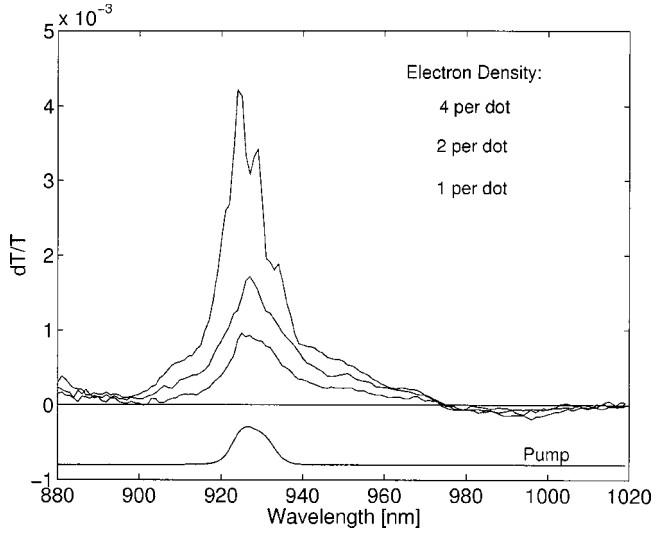

FIG. 2. Differential transmission spectra taken with different carrier densities at a delay of 100 fs. The spectra correspond to about one, two, and four electron–hole pairs per dot (bottom to top). The pump spectrum is shown on the bottom of the figure.

ground-state band consists of four levels each with a spin degeneracy of two. Photoluminescence data on this sample confirm that the excited-state interband transition (E2H2) is centered at 920 nm while the ground-state transition (E1H1) is centered near 980 nm.[6]

The pump–probe DT measurements are carried out at 10 K with two single-filament white-light sources for the pump and the probe pulses. The white-light sources are generated using a 100 fs 250 kHz regenerative amplifier output.[6] A 10 nm bandpass filter centered at 920 nm is used to select the pump pulse. For the probe pulse, we select the spectral band between 900 and 1020 nm with a RG1000 Schott filter and 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 resonantly generate carriers in the excited ($n=2$) states of the dot, and the differential transmission of the variably delayed probe pulse measures the transient occupation of the dot levels. The pump is chopped at 2 kHz, and the probe DT signal is detected with a lock-in amplifier and spectrally resolved to a 1 nm resolution.

In our first experiment, DT spectra are taken at different delays to observe the spectral evolution of the carrier population. DT spectra from five different delays are shown in Fig. 1 along with the pump spectrum. The pump pulse fluence is kept low such that it generates a carrier density of less than one electron–hole pair per dot. The five sequential spectral scans show two prominent dynamic features. First, within just a few hundred femtoseconds of photoexcitation, there is an extremely fast filling of off-resonance quantum-dot states. Second, there is a slower, but still very fast, population of the inhomogeneously broadened ground state indicated by a growing DT signal around 970 nm. The second of these events is due to the carrier relaxation from the excited state to the ground state of the quantum dot as was reported earlier.[6] The initial continuous spreading in the energy spectrum indicates a rapid redistribution of the carriers among coupled excited states. In the event that the dots are not coupled, one would expect the carriers to undergo intradot relaxation and show an evolving DT spectrum with narrow E2H2 and E1H1 peaks of only the dots that are pumped resonantly. Instead, we see a continuous, rapid energy spread throughout the inhomogeneously broadened excited states, indicating interdot coupling. The oscillations seen in the DT spectrum around the pump wavelength are a result of interference between the probe and scattered pump light.

In the second experiment, the DT spectral scans are taken as a function of the carrier density at a fixed delay of 100 fs. The injected carrier density is estimated by considering the amount of incident pump light that is absorbed by the dots. Since we do not know precisely the absorption coefficient of the dots at 920 nm, we estimate the total absorption in two different ways. First, we directly measure the incident, reflected, and transmitted beams to extract the absorbed energy. Second, we measure the DT signal near saturation and directly determine the absorption coefficient. Both of these techniques reveal the total absorption to be about 0.5%. With this value, we directly calculate the number of photons that is absorbed by the dots in the four layers to estimate the carrier density. Shown in Fig. 2 are three spectral scans with increasing carrier densities of approximately 1, 2, and 4 electron–hole pairs per dot. The overall magnitude of the DT signal rises with increasing carrier density, but the extent of the energy spread remains constant. In addition, the general asymmetric shape and proportions of the spectral redistribution remain the same. From this, we rule out carrier–carrier scattering as a mechanism for this rapid spectral spread.

We also investigate interdot tunneling theoretically by calculating the tunneling rate between two coupled dots. We first calculate the strain distribution in a vertically coupled two-dot system using the valence force field (VFF) model.[11,12] The values for bond-bending and bond-stretching parameters as well as the deformation potentials are taken from Ref. 13, which describes the application of the VFF model to self-assembled quantum dots. The virtual crystal approximation is used to determine these values for $In_{0.4}Ga_{0.6}As$. We then use a simple scalar effective-mass ap-

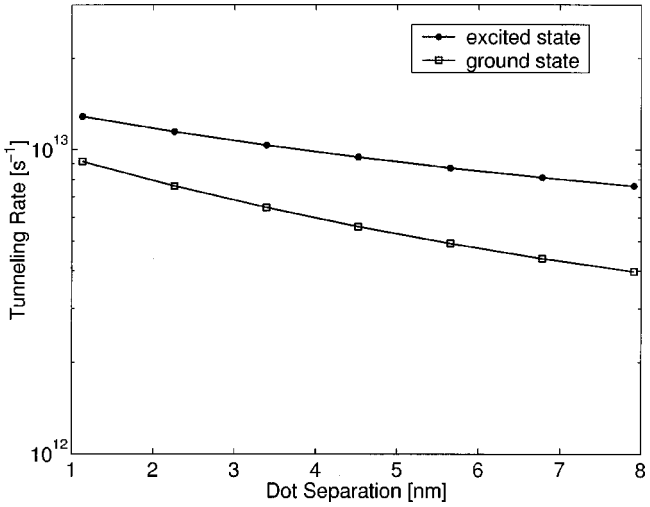

FIG. 3. Calculated tunneling rates for the ground and first-excited states as a function of interdot separation.

proximation to calculate the energies of the two-dot system by solving the Hamiltonian

$$\left(\frac{p^2}{2m^*}+V(r)+H_{\mathrm{str}}\right)\psi_n(r)=E_n\psi_n(r), \tag{1}$$

where $V(r)$ is the term due to the conduction-band offset and H_{str} is the strain contribution. We solve the eigenvalue problem numerically using a finite-difference approach with a mesh size of 5.65 Å, the GaAs lattice constant.

As the separation between the dots decreases, each state in the uncoupled system splits into two due to the coupling, with an energy difference ΔE varying exponentially with the interdot distance. We then determine the dot-to-dot tunneling rate Γ using the relation $\Gamma=\Delta E/2\hbar$.

Figure 3 shows the calculated tunneling rate for the ground and first-excited states as a function of interdot separation. The dot separation of our sample is 2.5 nm, for which we calculate the tunneling time in the excited state to be approximately 100 fs. This agrees reasonably well with the experimental results. Reference 14 describes the calculation formalism and results in greater detail. Similar calculations on lateral interdot coupling show that the in-plane dot-to-dot tunneling rate is much lower.[14] In addition, calculations of hole wave functions indicate that holes of low-lying energies are well localized in a deep potential, making tunneling of holes unlikely.[15] We conclude from this, the above calculations, and the experimental data, that the interdot coupling is not dominated by carrier–carrier scattering, as is the case in the carrier thermalization process in quantum wells, but by tunneling of electrons among the excited levels of closely neighboring vertically stacked quantum dots. This is in direct contrast to the model of isolated dots. In devices, such a coupling could lead to carrier dynamics exhibiting signs of phonon bottlenecks as electrons tunnel into dots without holes and prevent relaxation via an Auger-type scattering within the dot.

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