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 and as a result does not limit laser operation. 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. 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. Therefore, one might expect coherent coupling of the electronic levels to occur, 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 $\delta$-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, 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.

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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
between the pump pulse. For the probe pulse, we select the spectral band
centered at 920 nm while the ground-state transition
is centered near 980 nm.\(^6\)

In our first experiment, DT spectra are taken at different
delays to observe the spectral evolution of the carrier popu-
lation. DT spectra from five different delays are shown in
Fig. 1 along with the pump spectrum. The pump pulse flu-
ce 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, pop-
ulation of the inhomogeneously broadened ground state indi-
cated 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 spec-
trum 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 inter-
ference 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 consider-
ing the amount of incident pump light that is absorbed by the
dots. Since we do not know precisely the absorption coeffi-
cient of these dots at 920 nm, we estimate the total absorption
in two different ways. First, we directly measure the inci-
dent, 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 redistribu-
tion 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-
proximation to calculate the energies of the two-dot system by solving the Hamiltonian

$$\left( \frac{p^2}{2m} + V(r) + H_{st} \right) \psi_n(r) = E_n \psi_n(r),$$

where $V(r)$ is the term due to the conduction-band offset and $H_{st}$ 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 $\Delta E$ varying exponentially with the interdot distance. We then determine the dot-to-dot tunneling rate $\Gamma$ 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.

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