

Excited-state dynamics and carrier capture in InGaAs/GaAs quantum dots

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Subpicosecond time-resolved photoluminescence upconversion is used to measure the 12 K first-excited-state dynamics in large InGaAs/GaAs self-assembled quantum dots designed for 1.3 μm diode lasers. A comparison with the ground-state dynamics suggests that energy relaxation occurs in a cascade through the multiple discrete levels with an average interlevel relaxation time of ~ 250 fs. Excited-state emission is observed from two distinct populations. Due to the ultrafast relaxation from the excited state to the ground state in dots containing only a single exciton, the excited-state emission is dominated by the fraction of dots that capture more than one electron-hole pair. In this case, state filling in the ground state blocks the ultrafast relaxation channel, thereby enhancing the excited-state emission. While state filling and a random capture process dictate the primary features of the excited-state emission, at low excitation levels we find that the rise time of emission from the excited state is influenced by the much denser population of singly occupied dots.
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The delta-function-like density of states in self-assembled, quasi-zero-dimensional, InGaAs quantum dots (QDs) offer superior performance relative to quantum wells as active regions in laser diodes.¹⁻⁴ Compared with planar quantum-well lasers, QD lasers have exhibited lower threshold current and less temperature sensitivity.⁵⁻⁷ However, the discrete density of states may also impose impediments to carrier energy relaxation by optical phonon emission. This so called *phonon bottleneck*⁸⁻¹¹ which was predicted by Bockelman *et al.*¹² and Benisty *et al.*,¹³ could restrict the modulation rate of QD lasers, thus limiting their use for high-speed applications. On the other hand, reported relaxation rates vary significantly¹⁴ leaving the existence of significant phonon bottleneck effects an open question and leading to a variety of proposed models for the relaxation. Different rates reported for a range of QD structures and for a variety of experimental conditions suggest that the relaxation rate is strongly dependent on the details of the QD electronic structure, as confirmed by recent measurements on size-controlled QDs.¹⁵ Still, a quantitative and general understanding of the carrier relaxation is incomplete, and progress toward this understanding is complicated by the random nature of the process by which carriers in the wetting layer are captured by the QDs.¹⁶

Here, we expand on our prior studies of the ground-state dynamics in InGaAs/GaAs self-assembled QDs^{15,17} by investigating the excited-state dynamics as well. Measurements with better than 200 fs resolution were conducted over a broad range of injected carrier densities, from an average of less than one exciton per QD to many excitons per QD. Our results indicate an ultrafast cascade of carriers through the

multiple energy levels in these large QDs, with an average interlevel relaxation time of ~ 250 fs. The measured dynamics of carriers in the first-excited state are found to be influenced by the random nature of the carrier capture process, which leads to a finite probability of multiple excitation of the QDs, even at excitation levels resulting in an average of much less than one exciton per dot.¹⁶

The InGaAs/GaAs self-assembled QD sample was grown by a cycled submonolayer molecular-beam epitaxy approach. Two QD layers were formed from a deposition of 10 monolayers of $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ average composition. The two QD layers are embedded in a GaAs matrix, which is sandwiched between two AlAs barriers separated by 2000 Å. The QDs have a density of $\sim 10^{10}$ cm^{-2} , a lateral size of ~ 350 Å, and a height of ~ 110 Å after covering with GaAs. Although the alloy composition during growth is 50%, the center of the dots is indium rich, which evidently results in nearly parabolic potential wells.¹⁸ This is consistent with the nearly constant energy separation (~ 65 meV) between adjacent emission peaks, as evidenced by the continuous-wave photoluminescence (PL) [see Ref. 15 Fig. 1(a)].

Time-resolved PL upconversion of the QD PL was performed using ~ 120 fs pulses from a Kerr-lens mode-locked Ti:Sapphire laser tuned to 870 nm so that carriers are generated below the GaAs band edge into the wetting layer. The resulting PL was collected and both the ground-state and the first-excited state emission were upconverted in a 0.5-mm-thick LiIO_3 crystal. The upconverted signal was directed into a monochromator prior to detection by a cooled photomultiplier tube and a photon counter to provide a spectral resolution of ~ 5 meV. All measurements were conducted at a lattice temperature of 12 K to essentially eliminate thermal occupation of the QD excited states. We note that, since the excitation is in the wetting layer, separate capture of elec-

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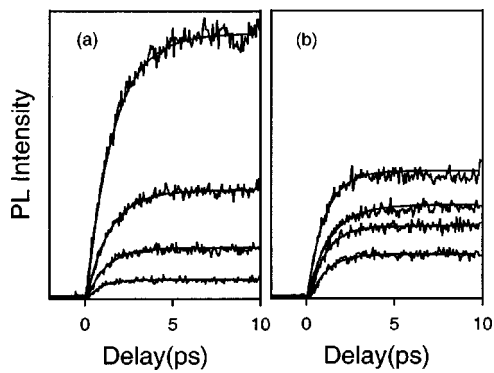


FIG. 1. Time-resolved PL for the first-excited state (a) and the ground state (b) at 12 K. The excitations for both states are $0.47 \mu\text{J}/\text{cm}^2$, $0.95 \mu\text{J}/\text{cm}^2$, $2.84 \mu\text{J}/\text{cm}^2$, and $9.45 \mu\text{J}/\text{cm}^2$, respectively. The solid lines are mono-exponential fits.

trons and holes can occur, but PL results only from QDs that have captured at least one electron-hole pair.

Immediately after excitation, the PL from both the ground state and the first-excited state increases rapidly with time in a manner that is well described by a mono-exponential rise to a constant level. Figure 1 shows data and mono-exponential fits for both the ground state and the first-excited state for several excitation fluences. We estimate that $1 \mu\text{J}/\text{cm}^2$ corresponds to an average of approximately one electron-hole pair per dot. We find that the emission associated with both the ground state and first-excited state reaches a quasi-equilibrium on a time scale of ~ 1 ps. Given that we observe transitions corresponding to at least four discrete energy levels, this rise time indicates an average interlevel relaxation time of ~ 250 fs. Such fast relaxation, which occurs at all excitation densities, indicates that, in these QDs, the phonon bottleneck is not significant. The data also illustrate that, at low excitation, the ground-state emission is much more intense than the excited-state emission. However, with increasing excitation, the PL intensity for the first-excited state begins to dominate, as would be expected due to the higher degeneracy for the excited state and state filling in the ground state. The dependence of the PL intensity on excitation for both transitions is quantified in Fig. 2. These results

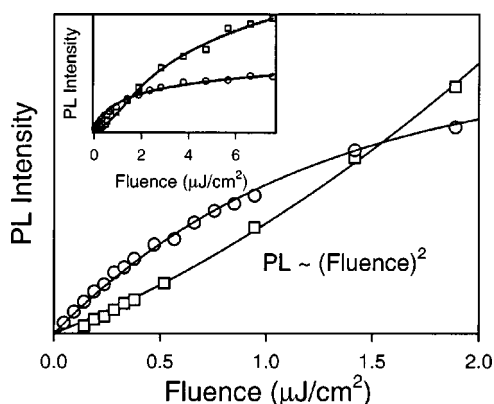


FIG. 2. Time-resolved PL intensity vs excitation fluence for the first-excited state (square) and the ground state (dot) 10 ps after zero at 12 K with the inset showing the same thing for the entire excitation range investigated. The solid lines are fits to the data. Note that the fit for the first-excited state at a low-excitation is quadratic, which indicates the first-excited state emission arises mainly from doubly occupied dots.

clearly indicate that first-excited-state emission is observed even at excitation densities well below the level required for state filling in the ground state, i.e., for less than one exciton per QD on average. For excitation levels well below that required for ground-state filling, the ground-state emission, as expected, increases linearly with excitation but the excited-state emission increases quadratically. This behavior suggests that the measured excited-state emission is dominated by those QDs that have randomly captured two excitons.

Measurements of the PL decay were also conducted to evaluate the lifetimes of the ground state and first-excited state. These data (not shown) display an exponential decay, and mono-exponential fits yield an excited-state lifetime of 230 ps and a ground-state lifetime of 460 ps. It is important to note that there is no significant density dependence to these lifetimes. That is, even when the excitation level is well below an average of one electron-hole pair per dot, the excited-state lifetime, which includes both radiative decay and relaxation to the ground state, is ~ 230 ps. This lifetime is more than two orders of magnitude longer than the rise time of the unsaturated ground-state emission. This indicates that the measured excited-state emission must arise primarily from dots in which relaxation into the ground state is blocked.

The rapid rise times associated with the ground state emission, the comparatively long lifetime observed for the first-excited state, even for an average excitation of less than one exciton per dot, and the density dependence of the excited-state emission all suggest that the measured excited-state PL arises primarily from a random population of QDs that have captured more than a single electron-hole pair. Due to the random capture process, such dots will be present at all excitation levels. In these dots, one electron-hole pair will rapidly relax into a ground state exciton, and Pauli blocking in the ground state will force (for appropriate spin) the second exciton into the excited state. Under such circumstances, carrier relaxation out of the excited state can occur through two paths: radiative recombination and scattering into the ground state after radiative decay of the ground-state exciton. The fact that the excited-state lifetime is half that of the ground state indicates that the radiative lifetime of the excited state is essentially equal to that of the ground state.

For dots that capture only a single electron-hole pair, electron occupation of the first-excited state is brief, since the ~ 1 ps rise time of the ground-state emission implies that most of those electrons quickly scatter into the ground state. In the absence of Pauli blocking in the ground state, the short lifetime of the excited state, as well as the weak oscillator strengths for recombination with all but the first-excited hole state, lead to a low probability per dot for excited-state emission.

Although most of our observations related to the excited-state emission can be explained by the statistical probability of multiple exciton capture in the QDs together with ground-state filling, a quantitative examination of the ground-state and excited-state PL rise times indicates that this alone can not fully describe our data. Figure 3 illustrates the excitation density dependence of the rise times for the ground state and the first-excited state. These data illustrate that at excitation

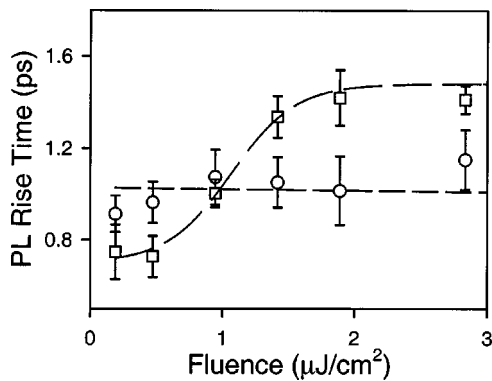


FIG. 3. PL rise time vs excitation fluence for the first-excited state (square) and the ground state (dot) at 12 K.

levels for which ground-state filling is complete (see Fig. 2), the excited-state PL rise time is 300–400 fs slower than that of the ground state. This is consistent with the model of random capture and state filling and is similar to observations described by Yuan *et al.*¹⁹ However, for excitation levels well below that required for complete filling of the ensemble ground state, the excited-state rise time is 200–300 fs *faster* than that of the ground state. A faster rise time for the excited-state emission is inconsistent with a state-filling model for the emission, but it is what would be expected for singly excited QDs in which emission occurs as carriers pass through the first-excited state. However, this mechanism would not result in the 230 ps lifetime measured for the excited state.

The apparent contradictory behavior of the low-excitation excited-state dynamics can be explained by recognizing that two populations of QDs contribute to the excited-state signal, i.e., a large population of QDs occupied by single excitons and a much smaller group of QDs randomly occupied by multiple excitons. While the excited-state transition probability is small for the singly occupied QDs, their large density results in measurable emission from the excited state. This emission is characterized by a rise time faster than that measured for the ground state and an extremely short lifetime that is characteristic of scattering into the unoccupied ground state. On the other hand, state filling in the dots with multiple excitons greatly increases the excited-state radiative transition probability by blocking scattering into the ground state. This results in measurable excited-state emission from this population, even though its density may be much smaller than that of the singly excited QDs. This emission is characterized by a rise time that is somewhat slower than that of the ground state, as ground-state occupation occurs prior to the excited-state emission, and a long lifetime controlled by radiative recombination. The relative excited-state lifetimes and the densities of the two populations control the measured dynamics, and a rate equation analysis of the emission suggests that the behavior exhibited in Fig. 3 can be readily obtained.

In summary, we have time resolved the ground-state and first-excited-state emission in large ($300 \text{ \AA} \times 150 \text{ \AA}$) InGaAs/GaAs QDs. The data indicate that even at low densities, such that on average there exists less than one exciton per QD, the most prominent features of the excited-state emission are dominated by the small group of QDs that have captured more than one exciton. On the other hand, for sufficiently low-excitation levels, excited-state emission from QDs occupied by single excitons does influence the emission rise time. The data are fully consistent with an ultrafast cascade of carriers through the multiple discrete levels in the QDs and with the random capture model proposed by Grundmann and Bimberg.¹⁶ Furthermore, we see no evidence of a significant phonon bottleneck in these structures.

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