Ultrafast carrier activation in resonantly excited 1.3 μm InAs/GaAs quantum dots at room temperature

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Carrier activation dynamics is measured in self-assembled InAs/GaAs quantum dots with a high degree of electronic state symmetry, at room temperature and following resonant excitation in the ground state. Carriers are activated to the first excited state on a 15-ps time scale in the low-excitation regime, and the total activation rate increases quadratically with the fractional dot occupation. Electron-hole interaction is identified as the dominant mechanism of electron scattering within the lowest confined states of a single quantum dot, circumventing the observation of a phonon bottleneck.

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Semiconductor quantum dots (QDs) are the perfect laboratory system to study carrier-carrier and carrier-phonon interaction in zero-dimensional solid-state systems. The fully discretized electronic density of states has a strong influence on the carrier dynamics, because the carrier energy relaxation within the discrete states of a single quantum dot is affected by the shape of the e-h wave functions and the nature of the phonon modes, and by the energy separation between the QD electronic states.1 In self-assembled QDs, it has been shown that electron energy relaxation is mainly mediated by Auger-type processes when carriers are injected in the barriers of the QDs at high intensities,2 while intradot relaxation (between the QD low-energy states) can be determined by a complicated interplay between phonon scattering3,4 and intradot3 or plasma Coulomb scattering.5 At low temperatures (<40 K), the observation of a “phonon bottleneck” has been reported in resonant-excitation experiments where carriers are created in the QD first excited state,7 but it has also been observed that electron-hole (e-h) scattering within a single quantum dot can influence the electron energy relaxation from the first excited state to the ground state.5 At higher temperatures, multi-LO-phonon or multphonon scattering involving LA phonons can effectively circumvent the single-LO-phonon bottleneck.6,7 To the best of our knowledge, the interplay between e-h and multiphonon scattering in the QD carrier dynamics at high temperatures has not yet been studied experimentally. Electron-hole scattering might be important up to room temperature in high in-plane symmetry QDs, where the high electronic symmetry results in a small e-h dipole moment and thus the exciton-LO-phonon interaction is low.9

In this paper, we show that electron-hole interaction within a quantum dot is likely to be the dominant mechanism of electron scattering between the lowest electronic levels of self-assembled InAs QDs at room temperature and for fractional dot occupations as low as 0.1, when the carrier dynamics is independent of excitation intensity. Our QDs feature a high level of electronic symmetry in polarization-resolved optical bleaching experiments. We adopt a scheme of resonant excitation in the ground state. Owing to the minimum number of e-h ground states, such an excitation scheme allows us to best control the excitation density and the initial conditions of the carriers. This scheme also restricts the number of possible scattering processes that can take place, due to energy conservation requirements.

The self-assembled InAs/GaAs QDs are grown by molecular-beam epitaxy on GaAs. The sample we investigated consists of four stacks of two QD layers each. Each QD layer consists of a InAs seed layer followed by a six-period (1–0.5 ML) GaAs-InAs superlattice and is designed for the fundamental optical resonance of the QDs to occur near 1.3 μm at room temperature.10 The QD layers within a stack are separated by 400 Å of GaAs, and adjacent stacks are separated by ~1500 Å of GaAs. From cross-section transmission electron microscopy, we estimate the QD density (Ndot) to be ~3–4×1010 cm−2 per layer. The QDs appear to have a base length <30 nm and a height <10 nm. Differential transmission (∆T/T) measurements are performed using 150-fs pulses from an optical parametric oscillator (pumped by a Ti sapphire laser), tuned to the QD fundamental optical resonance, in a standard degenerate pump-probe configuration with variable field polarizations, at near normal incidence on the sample. High signal-to-noise ratio is achieved by combining balanced detection with high-frequency modulation (~1 MHz) and lock-in detection. Time-resolved photoluminescence (PL) traces and spectra are obtained using a standard up-conversion technique. PL is excited by 150-fs pulses with variable field polarization, and the emission is collected by a Cassegrain reflector, focused onto a 1-mm-thick LiIO3, nonlinear crystal, up-converted using a residual beam from the Ti sapphire laser, then dispersed in a double spectrometer and detected by a low-noise GaAs photomultiplier. The excitation spot size (1/e2 diameter) used in the pump-probe (PL) experiment is approximately 70 (60) μm. All measurements are performed at room temperature.

Our high-quality QDs are characterized by the continuous-wave PL spectroscopy technique with excitation in the GaAs barriers. Spectra for different excitation densities are shown in Fig. 1. At high intensities, we observe three or more QD (interband) optical resonances (n = 1,2,3), as well as emission from the underlying InAs wetting layer and from the GaAs barriers. The number of QD transitions and their energy separations are qualitatively consistent with recent calculations based on an atomistic pseudopotential approach.11
Polarization-resolved $\Delta T/T$ measurements with resonant excitation at the QD fundamental ($n=1$) resonance are shown in Fig. 2. Panel (a) depicts the early $\Delta T/T$ dynamics for different pump and probe polarizations. For cross-linearly polarized ($\sigma^+\sigma^-$) pump and probe pulses, the signal decreases monotonically with increasing delay due to progressive recovery of absorption after pump bleaching. The overall dynamics exhibits a three-component exponential decay, the fastest component [visible in Fig. 2(a)] has a decay time of $\sim 0.7$ ps and is attributed to hole thermal activation, possibly via multiphonon absorption.\textsuperscript{12} The observed initial bleaching is maximum (minimum) for co(cross)-circular ($\pi^\pm\pi^\pm(\pm)$) polarizations, from which we deduce that the electronic ground states have a high degree of circular symmetry: The optically active e-h pair states have a quantum-well-like character, with total angular momenta $J_z = \pm 1$ (z is the growth direction) approximately.\textsuperscript{13} A quantitative analysis of the data shows that 90% or more of the carriers photoexcited by a $\sigma^+$ pulse are created in a given optically active e-h pair state ($J_z = -1$) and the remaining carriers are created in the orthogonal state ($J_z = +1$). The increase in bleaching with delay in the first 3 ps after excitation for $\sigma^+\sigma^-$ polarizations indicates that carriers undergo ultrafast spin-relaxation processes between the two optically active e-h states.\textsuperscript{14} A detailed study of spin-flip processes will be reported elsewhere. Figure 2(c) shows that the initial $\Delta T/T$ signal for different laser wavelengths reproduces the $n=1$ PL resonance spectrum, hence demonstrating that the signal originates from the dots.

The initial $\Delta T/T$ signal exhibits a saturation behavior versus pump intensity [Fig. 2(b)], with a fit saturation intensity ($P_{sat}$) of $\approx 0.9$ kW/cm$^2$ for $\sigma^+$-polarized pump pulses (empty dots). The high-intensity signal limit ($\Delta T/T_{max}$) is a measure of the linear absorption of the QD layer ($\Delta T/T_{max}$) in the graph.

FIG. 1. Cw photoluminescence (PL) spectra of the quantum dots with excitation in the GaAs barriers, for different excitation densities (in kW/cm$^2$). The diagram depicts the confined electron and hole states and the basic optical transitions of the quantum dots. According to the theoretical findings in Ref. 11, the e2-h3 optical transition is also allowed and contributes to the peak designated as $n=2$ in the graph.

FIG. 2. (a) Differential transmission ($\Delta T/T$) versus pump-probe delay for three different pump-probe field polarization configurations: co(cross)-circular ($\sigma^+\pi\sigma^+\pi$) and cross-linear ($\sigma^+\sigma^-$) polarizations. The excitation is resonant with the fundamental resonance absorption peak and the pump intensity is 0.5 kW/cm$^2$. The linear polarization of the pump is along the [110] crystallographic direction of GaAs. The solid lines are multicomponent exponential decay fits. (b) Initial (zero time delay) $\Delta T/T$ signal at the fundamental resonance peak versus pump intensity, for cross-linear (full dots) and cocircular (empty dots) polarizations. Solid lines are fits of the function $\Delta T/T(P) = \Delta T/T_{max}P/P_{sat}/(1 + P/P_{sat})$ to the data. (c) Initial $\Delta T/T$ signal for different central wavelengths of the laser pulses (big full dots), normalized in intensity to a cw PL spectrum (small full dots), for cross-linear polarizations. Very similar results are obtained with circular polarizations. The laser spectral width full width at half maximum is $\sim 11$ nm. $\Delta T/T_{max} = N_L\alpha_0 d$, where $N_L$ is the number of QD layers, $\alpha_0$ is the QD peak linear absorption coefficient, and $d$ is the QD layer thickness. We find $\alpha_0 d = 4 \times 10^{-4}$, in very good agreement with a previous experimental determination.\textsuperscript{15} The measurement of the saturation intensity also provides an accurate and unambiguous calibration of the carrier density versus excitation intensity. Since a linearly polarized field excites the two optically active e-h pair states almost equally, for such a pump polarization the saturation intensity is twice as large, and, in fact, the corresponding initial bleaching increases almost linearly with increasing pump intensity (full dots), up to the maximum accessible intensity. At saturation, i.e., for $P = 2P_{sat}$, the fractional dot occupation $\eta$ is equal to 0.25 (0.5) for a circularly (linearly) polarized pump pulses. A polarization selective (circularly polarized) pulse can create only one e-h pair in a given dot, while a polarization unselective (linearly polarized) pulse can create two e-h pair excitations.

In order to study the carrier activation dynamics from the ground ($n=1$) to the first excited ($n=2$) state, we measure the time-resolved PL signal at the QD $n=2$ resonance, following resonant excitation at the $n=1$ resonance. The main panel of Fig. 3 shows a PL spectrum of the QDs upconverted after PL activation completion (full dots). As a control experiment, we verify that the PL signal detected at the $n=2$ resonance peak for different excitation wavelengths reproduces the $n=1$ resonance spectrum (hollow diamonds).
This demonstrates that the $n=2$ PL is activated directly by 
\(e-h\) pairs scattered from the $n=1$ to the $n=2$ states and the 
contribution of the two-photon absorption in the GaAs barriers 
is completely ruled out for $P \leq 1.1$ kW/cm\(^2\). Excited-state PL time profiles are shown in the upper inset for different 
intensities, and an exponential rise function is fitted to the 
data. Since the holes undergo very fast thermal activation 
and the PL emission at the $n=2$ resonance occurs only 
provided that both an electron and a hole are simultaneously 
present in the respective $n=2$ states, the fit rise time is taken 
to be a direct measure of the electron activation time ($\tau_{\text{act}}$).

The activation rate ($1/\tau_{\text{act}}$) decreases with increasing 
intensity (Fig. 4) and for $P < 1.5$ kW/cm\(^2\) attains a constant 
value, which corresponds to an electron activation time $\tau_{\text{act}}$ = 14 ps. 
Based upon the above-mentioned control experiment, in the low-intensity limit we exclude any plasma effects 
due to carriers directly excited in the GaAs barriers,\(^6\) 
and thus the intensity independent $n=1$ to $n=2$ activation 
mechanism must rely on either electron thermal activation 
(possibly due to multiphonon absorption) or electron scattering 
by thermally excited holes within a single quantum dot.\(^5\) 
The former process is expected to yield a total PL rate $\Gamma_1$ proportional 
to the number dots $N_1$ where at least one $e-h$ pair has been injected ($\Gamma_1 = N_1/\tau_{\text{rad}}$, where $\tau_{\text{rad}}$ is the carrier 
radiative lifetime and $N_1 = 2 \eta N_{\text{QD}}$). The latter process is 
much more likely in the presence of two holes, i.e., in the 
dots where a linearly polarized pulse has injected two $e-h$ 
pairs, in view of the energy conservation requirements in the 
scattering process. A linearly polarized pulse has almost the 
same probability (equal to the fractional occupation $\eta$) of 
creating an $e-h$ pair in either optically active $e-h$ pair state; 

\[ \frac{1}{\tau_{\text{act}}} = \frac{\gamma_1}{\tau_{\text{rad}}} + \frac{1}{\tau_{\text{thermal}}} \]

therefore the total number of dots with two photoexcited $e-h$ 
pairs is $N_2 = \eta^2 N_{\text{QD}}$, which yields a total $e-h$ scattering rate 
$\Gamma_2 = N_2/\tau_{\text{rad}}$.

The inset of Fig. 4 shows the $n=2$ peak PL signal (times 
the measured PL decay time $\tau_{\text{PL}} = 500$ ps, which is assumed 
to be equal to $\tau_{\text{rad}}$) as a function of the fractional occupation $\eta$, 
in the low-intensity regime only, where two-photon absorption 
does not take place. Data are consistent with a quadratic 
power dependence, from which we infer that in the 
low-intensity limit $n=1$ to $n=2$ electron activation occurs 
mostly in the dots with two photoinjected $e-h$ pairs, and 
hence is mediated by $e-h$ scattering following hole thermal 
activation. The very fast characteristic time of 14 ps is of the 
correct order of magnitude for this process, given a hole 
thermal activation time $\approx 0.7$ ps and the discussion 
conducted in Ref. 5.

We finally point out that for circularly polarized excitation 
the signal magnitude is about a factor of 2–3 smaller than for 
linearly polarized excitation, hence making signal levels in 
the low-intensity regime too small for reliable results. However, 
this is consistent with our model interpretation since a 
circularly polarized pulse has a low probability of exciting 
two $e-h$ pairs in a given dot.\(^6\)

In summary, we have shown that carriers (electrons) resonantly 
 injected at room temperature in the ground state of 
self-assembled InAs/GaAs QDs, which feature a high degree 
of electronic state symmetry, are activated to the first excited 
state on an ultrafast (15-ps) time scale, for small fractional 
dot occupations, in a regime where the scattering time is 
pump intensity independent. The overall phenomenology 
strongly suggests that electron activation takes place in the 
dots where two $e-h$ pairs are created and is mediated by 
electron-hole scattering following ultrafast hole thermal 
activation. In conclusion, at room temperature the phonon 
bottleneck is always circumvented in self-assembled InAs- 
GaAs QDs, and electron-hole scattering can dominate over 
electron-multiphonon scattering in determining the carrier 
dynamics within the QD confined states.


12 F. Quochi *et al.*, Physica B (to be published).

13 This suggests that the dots are either disk or lens shaped and may result from our design of the QD layers.

14 Very recently, ultralong (carrier recombination limited) excitonic spin-relaxation times have been reported in pyramidal self-assembled QDs at low temperatures, M. Paillard *et al.*, Phys. Rev. Lett. 86, 1634 (2001).


16 For a 10% excitation “leak” into the $J_z = +1$ e-h pair state in the case of $\sigma^+$-polarized pump pulses, the $n=2$ PL intensity is expected to be about 30% of that excited by linearly polarized pulses, in good agreement with the experimental results.