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Intraband relaxation via polaron decay in InAs self-assembled quantum dots

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Three distinct channels for the decay of polarons in InAs quantum dots are identified using energy and temperature-dependent far-infrared pump-probe spectroscopy. For energies up to ~53 meV a monotonic increase of the polaron relaxation time is observed, the polaron decay being shown to occur into two longitudinal acoustic phonons. Above this energy additional decay channels into two optical phonons are allowed, leading to the observed reduction of the decay time. We also demonstrate interlevel polaron transfer between the closely spaced *p*-like excited states, with measured transfer times in good agreement with calculations for an acoustic-phonon-mediated process.

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Over the last decade carrier relaxation in quantum dots (QDs) has been a major topic of investigation in semiconductor physics. Early studies[1–3] considered electron relaxation via phonon emission in the weak-coupling regime. Because of the discrete density of electronic states in QDs and weak energy dispersion of the longitudinal optical (LO) phonons it was suggested that efficient electron relaxation (<1 ns) is only possible for energies <3 meV from the LO phonon energy,[3] commonly known as the phonon bottleneck effect. However, more recent studies[4–7] have shown that, due to the discrete nature of the confined states, electrons and phonons in InAs QDs are not in the weak-coupling regime but are strongly coupled to form polarons. This has two main consequences for intraband relaxation in QDs compared with bulk or quantum well systems. The first is that the decay time of polarons is determined by the instability of the LO-phonon component,[8–10] providing a lower limit of several picoseconds for the polaron decay time which is longer than the typical electron relaxation time in bulk or quantum well systems (~1 ps). Second, the phonon bottleneck effect is removed for polaron decay, with a relatively weak increase in the decay time predicted[10] over a wide energy detuning from the LO-phonon energy.

Magnetotransmission measurements[6,7] performed on *n*-type self-assembled InAs/GaAs QD structures have demonstrated clear evidence for the polaron picture with strong anticrossings observed close to multiple LO-phonon energies. Calculations of polaron decay times[8–10] indicate that the polaron lifetime is determined by the finite lifetime of the LO-phonon component, which is unstable due to phonon anharmonicity.[11] Assuming that the polaron decay channel is the same as the LO-phonon decay channel in bulk GaAs, i.e., 1LO→1LO+1 transverse acoustic (TA), Verzelen *et al.*[8] predicted that the efficient relaxation should only occur inside an energy window around the LO-phonon energy $(\hbar\omega_{LO})\pm 8$ meV, outside which the polaron decay is quenched. Jacak *et al.*[6] have compared this relaxation process with decay to 1LO+1 longitudinal acoustic (LA) phonon and Li *et al.*[10] have considered polaron decay into 2LA phonons (cubic overtone). These decay channels are expected to extend the energy range over which polaron decay can occur and in Ref. 10 has been shown to lead to the efficient relaxation of excited polarons even for quite large detunings (~20 meV) from the LO-phonon energy. Such large energy ranges for efficient relaxation from excited states are expected to lead to significant suppression of phonon bottleneck effects in quasi-zero-dimensional quantum dots, decay times in the range of 10–50 psec being found in the present work. The polaron decay model presented in Ref. 10 has been used to explain the experimental results from a previous report of polaron dynamics in self-assembled QDs, but without identification of the phonons involved in the final state.[12]

In the present communication we present the results of polaron lifetime studies for a series of samples over a wide energy range from close to the LO-phonon energy (40 meV) up to ~60 meV. Careful analysis of the temperature dependence of the polaron lifetime has allowed us to identify unambiguously the main polaron decay channel as cubic overtone to 2LA phonons. Using the approach developed by Li *et al.*,[10] we have extracted the low-temperature LO-phonon lifetime for InAs QDs, as well as the electron-phonon coupling strength. From our measurements we also identify an energy threshold (~53 meV) above which decay to optical phonons becomes significant. In addition, we have measured the polaron transfer time between the two lowest-energy excited states for a set of samples with different energy separations between the two states. We find very good agreement with our calculations assuming acoustic-phonon-assisted transfer between the states.

The InAs/GaAs QD samples were grown on (100) GaAs substrates by molecular-beam epitaxy in the Stranski–Krastranow mode. The studied structures contain either 50 or 80 layers of InAs QDs separated by 50-nm intrinsic GaAs spacers. Prior to growth of the multilayer samples, uncapped reference samples were grown from which we were able to

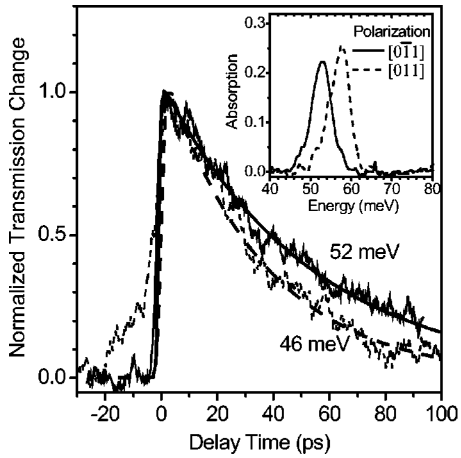

FIG. 1. Normalized 5 K transmission change as a function of time delay between the pump and probe for different pump (probe) energies of 46 meV (dashed line) and 52 meV (solid line). The numerical fits used to extract the decay times are indicated by solid lines. The inset shows a typical normal incidence absorption spectrum for incident radiation polarized along $[0\bar{1}1]$ (solid line) and [011] (dashed line) for a multilayer InAs/GaAs QD sample.

determine the QD density (typically $\sim 4 \times 10^{10}$ cm^{-2}) from atomic force microscope analysis. In order to achieve a population of $\leqslant 1$ electron per dot the multilayer samples were δ doped in the GaAs barrier 2-nm below the QD layers with a sheet density equal to the measured dot density.

Typical normal-incidence linear absorption spectra of the QD samples measured at 5 K are shown in the inset of Fig. 1. Incident radiation polarized along $[0\bar{1}1]$ ([011]) excites a transition from the *s*-like ground state (s) to the lower (higher) energy laterally confined *p*-like excited state. The absorption peaks associated with transitions to the lower-energy *p*-like state (p_-) and higher-energy *p*-like state (p_+) are centered at 53 and 58 meV, respectively, for this sample. The $\sim$5 meV splitting between the two peaks can be explained by elongation of the QDs along $[0\bar{1}1]$,[13] piezoelectric field effects,[14] and the atomistic symmetry.[15] All of the QD samples studied have generally similar absorption spectra, although some samples have a larger linewidth ($\sim$10 meV) due to a more inhomogeneous QD size distribution, allowing the study of polaron decay over a relatively large energy range in the same sample. By performing pump-probe spectroscopy on a range of samples containing QDs with varying lateral sizes we have been able to measure polaron dynamics over an energy range of $\sim$20 meV.

We have performed degenerate pump-probe measurements with the Dutch Free Electron Laser for Infrared Experiments (FELIX) facility in order to determine the polaron decay times with $\sim$1 ps time resolution using a three-beam pump-probe-reference technique similar to that reported in Ref. 16. Typical time evolutions of the probe signal following the arrival of the pump pulse (pump-probe signal) are shown in Fig. 1 for pump/probe energies of 46 and 52 meV, with incident radiation polarized along the $[0\bar{1}1]$ direction (i.e., excitation of the *p*-state). A clear energy dependence of the relaxation time is measured, increasing with increasing

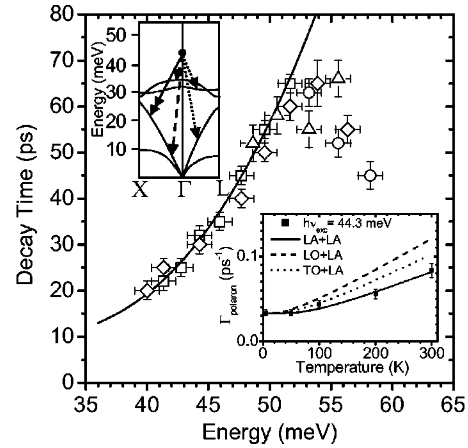

FIG. 2. Measured energy dependence of the decay time at 5 K (solid squares) and calculated polaron decay times (solid line) assuming an optical-phonon lifetime $\tau_{ph} = 13$ ps and electron-phonon coupling strength $g = 6$ meV. The upper inset shows a schematic GaAs phonon dispersion and three possible polaron decay channels, to 2LA phonons (solid line), 1LO+1LA (dashed line), and 1TO +1LA (dotted line). The lower inset shows the measured (solid squares) and calculated temperature-dependent polaron decay rates at 44 meV assuming decay to 2LA phonons (solid line), 1LO +1LA (dashed line), and 1TO+1LA (dotted line).

pump energy. In order to extract the transmission recovery times a monoexponential fitting of the experimental data has been performed for the range of samples investigated. As shown in Fig. 2, where the spectral dependence of extracted decay times for all samples is presented, the polaron lifetime increases monotonically with increasing energy up to $\sim$53 meV. The energetic dependence of the decay time is a clear signature of polaron decay, arising from the reduced phonon fraction of the polaron at larger energy detunings from the LO-phonon energy. The solid line in Fig. 2 is a best fit of the experimental data using the model presented in Ref. 10, which assumes polaron decay to 2 LA phonons. Very good agreement is obtained up to $\sim$53 meV using the parameters $\tau_{ph} = 13$ ps (optical-phonon lifetime at 36 meV) and $g = 6$ meV (electron coupling strength to all the LO-phonon modes).[17]

It should be noted that while the model of Ref. 10 fits the experimental data very well, it does not provide an unambiguous identification of the polaron decay channel. In this energy range (up to $\sim$53 meV) three main decay processes are possible, while satisfying energy and momentum conservation, involving either the creation of two equal-energy LA phonons or the creation of a higher-energy LO/TO phonon and a lower-energy LA phonon as indicated in the upper inset of Fig. 2. The decay channel can be determined by measuring the temperature dependence of the decay time as the polaron damping rate is proportional to the occupation numbers of phonons $n(\hbar\omega_i)$ involved in the decay.[18] Then, assuming the scattered energy is the polaron energy,

$$\Gamma_{polaron} = \gamma_0[1 + n(\hbar\omega_i) + n(\hbar\omega_j)], \tag{1}$$

where γ_0 is an effective anharmonic constant and $\hbar\omega_i$ ($\hbar\omega_j$) is the energy of phonon belonging to the i (j) branch, thus

$\hbar\omega_i + \hbar\omega_j = \hbar\omega_{polaron}$. The measured low-temperature relaxation rate fixes the only free parameter γ_0. As shown in the lower inset of Fig. 2, the temperature dependence of the polaron damping rate calculated for the LO/TO plus LA channels exhibit a stronger temperature dependence than for the cubic overtone decay channel (to 2LA phonons), due to the involvement of a lower-energy LA phonon in the former processes. Also shown in the lower inset of Fig. 2 is the measured temperature dependence of the polaron relaxation rate ($\Gamma_{polaron} = 1/\tau_{polaron}$) at a pump/probe energy of 44 meV. The very good agreement with the experimental data of the calculated damping rate for polaron decay to 2 equal-energy LA phonons clearly demonstrates that this decay channel dominates for energies up to $\sim$53 meV.

Above $\sim$53 meV a clear change in the energy dependence of the experimentally determined decay times occurs, with the measured lifetimes reducing from a maximum of $\sim$65 to $\sim$45 ps at 58 meV. This threshold behavior is well accounted for by the onset of a new two phonon decay mechanism, the possibility of multiple LO/TO-phonon decay. The energy threshold of $\sim$53 meV for these processes is consistent with a simple model based on bulk phonon energies. In this case, the lower limit for polaron decay is expected to occur close to twice the zone-edge InAs LO-phonon energy ($2 \times LO_{InAs} \sim 52$ meV) or twice the InAs TO-phonon energy ($2 \times TO_{InAs} \sim 55$ meV). The strong deviation of the measured decay times from those predicted using the model of Ref. 10 arises, because the model does not take into account these additional decay channels.

By performing pump-probe measurements using radiation linearly polarized along [011] we are also able to measure polaron decay from the p_+ excited state. The decay times we obtain can be directly compared to those for excitation of the p state (radiation polarized along $[0\bar{1}1]$) because the overlapping, inhomogeneously broadened s to p_-/p_+ absorption peaks allow pump-probe measurements to be performed at the same energy for either polarization. Polarons in the p_+ excited state may either decay directly to the ground state (s) or via the intermediate p_- state to s, meaning that any difference of the measured decay time for the two orthogonal polarizations is a result of polaron transfer from p_+ to p_-.

We have performed pump-probe measurements on three QD samples with p_--p_+ splittings (ΔE) varying from 3.7 to 5.5 meV. For each sample the pump-probe signal is measured at the same energy (indicated by the arrows in Fig. 3) with orthogonal linear polarizations. As shown in Fig. 3, for all samples we observe a shorter decay time for the p_+ state relative to the p_- state at the same energy. This is a result of the additional parallel decay channel for polarons in the p_+ state, involving relaxation to the ground state via the p_- state as shown schematically in the inset of Fig. 4. We can deduce the p_+ to p_- transfer time $\tau_{p_+ \to p_-}$ from the experimental data by fitting the pump-probe signal (solid line, Fig. 3) using the following combined rate equations:

$$\frac{dN_s}{dt} = -I\sigma(N_s - N_{p_+}) + \frac{N_{p_+}}{\tau_{p_+ \to s}} + \frac{N_{p_-}}{\tau_{p_- \to s}}, \qquad (2)$$

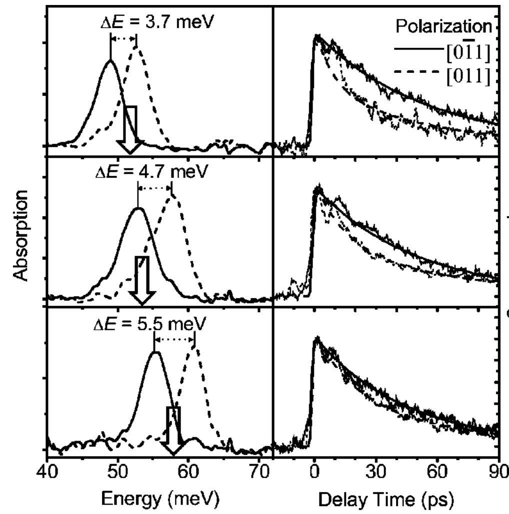

FIG. 3. Normal incidence absorption spectra (left-hand side) for three samples with varying p_--p_+ splittings and corresponding pump-probe curves (right-hand side) measured at the energies indicated by the arrows. The arrow width represents the spectral width of the pump (probe).

$$\frac{dN_{p_+}}{dt} = I\sigma(N_s - N_{p_+}) - \frac{N_{p_+}}{\tau_{p_+ \to s}} - \frac{N_{p_+}}{\tau_{p_+ \to p_-}}, \qquad (3)$$

$$\frac{dN_{p_-}}{dt} = \frac{N_{p_+}}{\tau_{p_+ \to p_-}} - \frac{N_{p_-}}{\tau_{p_- \to s}}, \qquad (4)$$

where I is the pump intensity and σ is the absorption coefficient of optical transitions from s to p_+ states. N_i is the population of the i state, where $i = s, p_-, p_+$. The decay time $\tau_{p_+ \to s}$ for direct transitions from p_+ to s states is assumed to be the same as for p_- at the same excitation energy. When pumping into p_+ at energy Ep_+, the $\tau_{p_- \to s}$ time is determined from the decay time measured separately at an energy Ep_+-ΔE. Following this procedure we obtain p_+-p_- times of 15, 20, and 35 ps for $\Delta E = 3.7$, 4.7, and 5.5 meV, respectively.

We have calculated the irreversible decay from the p_+ state to p_- via acoustic-phonon emission using the Fermi

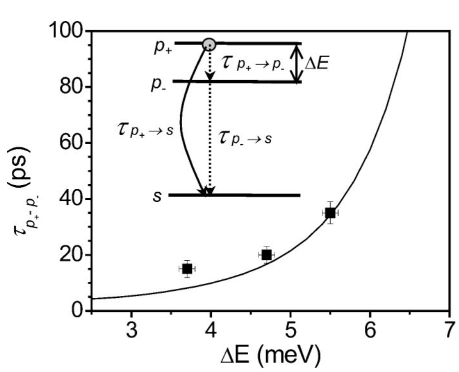

FIG. 4. Measured (solid circles) and calculated (line) p_+ to p_- transfer times. The inset shows the possible relaxation process for polarons from the p_+ state.

golden rule. The electronic states considered in the calculation differ very slightly from polaronic states as long as we are interested in computing the population transfer of the excited states in the large detuning limit. For the calculations we assume bulklike and isotropic acoustic phonons [$\omega(\vec{q}) = c_s q$, where c_s is the sound velocity in the medium, taken equal to that in GaAs]. The scattering time $\tau_{p_+ \to p_-}$ for the $p_+ \to p_-$ relaxation is given by

$$\frac{1}{\tau_{p_+ \to p_-}} = \frac{2\pi}{\hbar}[1 + n_{ac}(q_s)] \frac{|V_{DP}(q_s)|^2 q_s^2}{8\pi^3 \hbar c_s} \times \iint \sin\theta d\theta d\varphi |\langle p_+ | e^{-i\vec{q}.\vec{r}} | p_- \rangle|^2, \qquad (5)$$

$$\hbar c_s q_s = \varepsilon_{p_+} - \varepsilon_{p_-}, \qquad (6)$$

where θ and ϕ are the spherical angles of the wave vector $\mathbf{q_s}$ and $V_{DP}(\mathbf{q})$ is the deformation potential coupling term: $|V_{DP}(\mathbf{q})|^2 \propto \hbar c_s q$. The calculated scattering time is plotted (solid line) as a function of the energy separation between p_+ and p_- (ΔE) in Fig. 4. The increase of the scattering time with increasing ΔE arises mainly from the rapid decrease of the form factor in Eq. (5), with the dependence agreeing well in both magnitude and trend with our experimental results over the investigated range of ΔE.

In conclusion, we have observed three distinct decay mechanisms of polarons in InAs/GaAs self-assembled quantum dots. The first involves polaron decay to two longitudinal acoustic phonons which dominates below ~53 meV. Above this threshold energy a second process can occur, involving multiple optical-phonon decay. The third process involves polaron transfer between closely spaced (~5 meV) excited states, for which we find very good agreement between our experimental results and an acoustic-phonon-assisted transfer model.

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[17] We use the GaAs LO phonon energy of 36 meV to remain consistent with Ref. 10, however, our experimental data can also be well fitted assuming InAs LO phonons (energy ~30 meV) with only a small change in the coupling strength to ~6.5 meV. It is not possible for us to determine the precise nature of the LO phonon component of the polaron from the present measurements.

[18] A similar analysis has previously been applied to LO phonon decay in, for example, Ref. 11.