Tunneling carrier escape from InAs self-assembled quantum dots

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Deep-level transient spectroscopy measurements in InAs quantum dots (QDs) grown in both $n$-GaAs and $p$-GaAs show that tunneling is an important mechanism of carrier escape from the dots. The doping level in the barrier strongly affects the tunneling emission rates, enabling or preventing the detection of a transient capacitance signal from a given QD level. The relative intensity of this signal acquired with different rate windows allows the estimation of tunneling emission energies.

Stranski–Krastanow quantum dots (QDs) have recently attracted much attention due to their unique optical and electronic properties, which are enabling a wide range of novel applications. The transition energies between QD electron and hole levels can be directly measured from photoluminescence (PL) peaks. However, PL provides little information on electron and hole levels relative to the barrier band edges. Space charge techniques such as capacitance–voltage spectroscopy ($C–V$) and deep-level transient spectroscopy (DLTS) allow absolute positioning of the QD levels, providing complementary information to PL. Carrier capture and escape dynamics of the dots can also be studied by means of DLTS. There has been some discrepancy between different published works on DLTS in the extensively studied InAs/GaAs self-assembled QDs. Direct measurements of the energy difference between the dot levels and the energy band of the barrier have been reported, while other works show evidence of capture barriers into the dots. The possibility that some of the DLTS signals detected are originated from traps near the dots has also been suggested. To this day, only one study has reported clear detection of electron escape by means of tunneling in InAs QDs. In the present work, DLTS measurements performed on InAs QDs embedded in $n$- and $p$-GaAs show that tunneling is an important escape mechanism in quantum dots. Tunneling emission rates and energies are estimated by means of $C–V$ and DLTS.

QD structures were grown by molecular beam epitaxy. Two samples with eight 50-nm layers of $n$-type ($p$-type) GaAs ($n = p = 10^{18}$ cm$^{-3}$) terminated with InAs QDs ($\approx 2$ ML coverage) were grown over a 300-nm-thick, $n^+$-doped GaAs buffer layer. A final GaAs capping layer with the same doping level was deposited. A top Schottky diode and back ohmic contacts were formed for the $n$-type sample, and back and top ohmic contacts were formed on the $p$-type sample. The DLTS measurements were carried out at delay times $\tau$ in the (0.02–1000) ms range and at a rate window of 4.3$\times$ $\tau$.

Analysis of island sizes and densities using atomic force microscopy in air give average diameters of 40 nm, 5 nm heights, and concentrations of $3\times10^{10}$ cm$^{-2}$ for uncapped QDs grown under the same conditions. PL spectra for these structures were obtained at 300 and 77 K using an Ar$^+$ laser for excitation and a cooled Ge detector with lock-in techniques for signal detection. The extrapolated ground state emission at 4 K is $\sim 1.15$ eV, with full width at half maximum $\sim 130$ meV.

A typical $C–V$ profile obtained at 75 K for the $n$-type sample is shown in curve A (filled circles) of Fig. 1. Three plateaus corresponding to three different layers of dots are observed in addition to the background capacitance from the doped GaAs layers. Since only one plateau per QD level is expected, we attribute each plateau to the emptying of a single QD level. Curve C (filled circles) of Fig. 1 shows a similar behavior for the $p$-type sample. High leakage current for this sample allows biasing only to 2 V, therefore, only two plateaus were observed.

Models of different level of sophistication have been used to study the $C–V$ profile of single QD layers. A simpler analysis of the $C–V$ profile is possible by calculating the charge in the dots as a function of the applied voltage by using the expression reported in Refs. 3–5 for the density of electron states in the QD sheet but assuming that the...
depletion-region approximation still holds. The total capacitance is then given by the QD-related capacitance plus the background capacitance. This method, already applied using a lever-arm relation for a single layer of dots in Ref. 4, facilitates the calculation for a multilayered system. Within this approach we obtain the best fit (solid line) to curve A of Fig. 1 when one QD level with energy $E_n = 250 \text{ meV}$ below the GaAs conduction band and energy dispersion $\Delta E_p = 140 \text{ meV}$ is taken into account. For the $p$-type sample we obtain the best fit (solid line) to curve C with one single QD level with energy $E_p = 110 \text{ meV}$ above the GaAs valence band and energy dispersion $\Delta E_n = 140 \text{ meV}$.

Figure 2 (open circles) shows a DLTS spectrum for the $n$-type sample, consisting of a quasiflat signal that extends over the whole range of temperatures. A DLTS peak (not shown in the graph for scaling reasons) with an activation energy of $0.40 \text{ eV}$ appears for higher temperatures. Since this peak was only detected for positive or low negative filling biases, we believe it to be related to deep surface traps and not to the dots. The spectrum for the $p$-type sample (filled circles) shows a flat signal for the lower temperatures, and a step-like reduction to zero signal at about $55 \text{ K}$. To understand both spectra we must take into account that two carrier-escape mechanisms exist: thermal escape and tunneling. In the former, the quantum dot behaves like a deep trap and hence its thermal emission rate $e_{\text{th}}$ is given by $e_{\text{th}} = A T^2 \exp(-E_n/kT)$, where $E_n$ is the activation energy of the QD level and $A$ is a temperature-independent constant proportional to the capture cross section. For the next rate windows available, no step-like reduction is detected for the $n$-type sample as in Fig. 2 up to $325 \text{ K}$ because the DLTS signal detected at higher temperatures hinders any other feature. The inset of Fig. 2 plots the intensity of the flat signal at $20 \text{ K}$ for both the $n$-type and the $p$-type samples as a function of the delay time. For the $n$-type sample, the flat signal is detected with the lowest rate window available (delay time of $0.02 \text{ ms}$). For the next wider rate windows (delay times of $0.05$ and $0.1 \text{ ms}$) the flat signal detected is strongly reduced, which is a consequence of high rate of electron escape by tunneling. For the next rate windows available, no signal at all is detected. To further confirm that the flat signal obtained for the $n$-type sample is originated by electrons escaping from the dots, we have measured its intensity at $20 \text{ K}$ versus applied reverse bias. A positive filling bias was used to include the first layer of dots. The result, plotted in Fig. 1 (curve B), shows three plateaus that coincide with the plateaus of the $C-V$ measurement (curve A). The intensity of the flat signal increases when the applied reverse bias is sufficient to include a given layer of dots. For higher reverse bias the tunneling escape rate increases due to the increase of the electric field, and this diminishes the intensity of the flat signal.

For a carrier captured in a QD level to escape, the level must be above the Fermi energy in the barrier region. This happens for a certain onset reverse bias $V_c$ that, within the depletion approximation, satisfies the relation $-q\psi(V_c) + E_h = q(V_c + \psi_0) - E_F$, which leads to

$$E_h - q(V_c + \psi_0)\left(1 - \frac{L}{W}\right)^2 + E_F = 0,$$

where $\psi(V_c)$ is the potential at the dot layer, $E_F$ is the Fermi energy at the barrier, $W = [2(\psi_0 + V_c) e \epsilon_0 / (q N_{A,D})]^{1/2}$ is the depletion-region width, $L$ is the distance from the dots to the diode junction, $N_{A,D}$ is the doping level at the barrier and $\psi_0$ is the built-in potential. For this voltage $V_c$, the electric field $F$ at the dots can be calculated using the depletion approximation. By inserting the value of $F$ into Eq. (1), the lowest emission energy that gives rise to tunneling rates detectable with the shortest rate window available (delay time of $0.02 \text{ ms}$) can be calculated. A lower emission energy would yield carrier escape rates too fast to be detected with our instrumentation. We calculate that our lowest experimentally detectable emission energy is about $0.17 \text{ eV}$ for electrons,
about 0.15 eV for light holes, and about 0.060 eV for heavy holes. We have used the values \( m_e^* = 0.068 \), \( m_{lh}^* = 0.076 \) and \( m_{hh}^* = 0.50 \) for the effective mass of the different carriers.\(^1\)

Hence, the doping level in the \( n \)-type barrier does not allow detection of DLTS signals from shallower QD electron levels (higher-energy excited states measured by PL). For the \( p \)-type sample, we attribute the DLTS signal obtained to tunneling from a heavy hole level, since its emission energy is of about 0.10 eV, whereas no light hole levels lower than 0.15 eV can be observed for the experimental conditions of this work. Different results reported in the literature on InAs QDs could be accounted for by different doping levels in the GaAs matrix. Additional investigation on samples with different doping levels should be performed.

Finally, it is possible to evaluate the tunneling emission energy by measuring the relative intensity of the tunneling-related flat signal of two different DLTS spectra acquired with two different rate windows, defined by the time intervals \( (t_1, t_2) \) and \( (t'_1, t'_2) \), respectively.\(^10\)

The relative intensity of these tunneling signals is given by

\[
I = \frac{\exp(-e_{\text{tun}}t_2) - \exp(-e_{\text{tun}}t_1)}{\exp(-e_{\text{tun}}t'_2) - \exp(-e_{\text{tun}}t'_1)}.
\]

The emission energy can then be evaluated by using Eq. (1) with the value of \( e_{\text{tun}} \) obtained from Eq. (3). With this procedure, and using the data for the \( n \)-type sample plotted in the inset of Fig. 2, we estimate tunneling emission times for the \( n \)-type sample in the (20–50) \( \mu s \) range, which leads to an emission energy of 0.27 ± 0.04 eV which is close to the value obtained by \( C \)–\( V \). For the \( p \)-type sample, we estimate tunneling times between (30 and 500) \( \mu s \), which gives rise to an emission energy of 0.13 ± 0.03 eV, again, close to the value obtained with the \( C \)–\( V \) analysis. This value is also close to the value 0.10 ± 0.02 eV obtained for the activation energy using the position of the step-like reduction in the spectra of the \( p \)-type sample. Table I displays the different energy values for the electron (hole) levels relative to the GaAs conduction (valence) band obtained in this work. The proximity of energy values determined with different procedures seems to indicate that both the low-temperature flat signal and the step originate from the same QD level, in contrast to the two-level escape mechanism proposed in Ref. 7. Although Coulomb charging effects prevent the observation of higher excited states by space charge techniques,\(^13\) it is possible that two or even more QD levels close in energy are responsible for the \( C \)–\( V \) and DLTS spectra obtained here. In this case, the energy values that we calculate would be average values of the QD levels involved. Whereas \( C \)–\( V \) provides information about discharging of the QD levels with respect to the position of the Fermi energy, DLTS provides information about the barrier that the carriers must overcome to escape. It should be noted that the sum of the hole and electron energy levels determined with \( C \)–\( V \) and DLTS plus the ground-state recombination energy for the dots studied in this work 1.15 eV at 4 K) is close to the GaAs band gap 1.52 eV at 4 K). This fact and the closeness of the QD-level energies obtained by \( C \)–\( V \) and DLTS do not support the concept of capture barriers for the carriers into these QDs. This is in agreement with previous results on temperature-dependent PL\(^14\) and time-resolved PL experiments,\(^15\) in which important capture barriers have only been identified in low-density QDs, but not in high-density QDs like those studied here.

In conclusion, DLTS signals detected from InAs QDs are strongly affected by carrier escape by tunneling, with escape rates strongly dependent on the doping density of the barrier. The tunneling emission energies do not support the existence of high capture barriers into these QDs.

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\(^{1}\) D. Bimberg, M. Grundmann, and N. N. Ledentsov, Quantum Dot Heterostructures (Wiley, Chichester, 1998).


