Electric-field-dependent carrier capture and escape in self-assembled InAs/GaAs quantum dots

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Photoluminescence and complementary photocurrent spectroscopy, both as a function of electric field, are used to probe carrier capture and escape mechanisms in InAs/GaAs quantum dots. Carrier capture from the GaAs matrix is found to be highly field sensitive, being fully quenched in fields of only 15 kV/cm. For fields less than 20 kV/cm, carriers excited in the wetting layer are shown to be captured by the dots very effectively, whereas for fields in excess of 50 kV/cm tunnel escape from the wetting layer into the GaAs continuum is dominant. For excitation directly into the dots, radiative recombination dominates up to 100 kV/cm.
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InAs/GaAs self-assembled quantum dots are attracting widespread interest due both to their zero-dimensional properties,[1] and the potential they offer in device applications such as ultra-low-threshold lasers[1] and memory systems.[2] In many applications, electrically injected or photoexcited carriers are first created in the GaAs barriers prior to capture by the two-dimensional (2D) wetting layer (WL) and quantum dots. Relaxation to the quantum-dot ground state then occurs by LO phonon emission[3] or Auger scattering[4] on time scales of 1–50 ps.[5] Less information is available, however, concerning the initial capture of carriers from the GaAs continuum.[6] A further factor of importance is the escape of carriers from the quantum dots. Both thermally activated and tunneling escape have been identified,[7–9] but how they compete with relaxation and recombination is much less clear.

In this letter, we employ photoluminescence (PL) spectroscopy and photocurrent (PC) spectroscopy on InAs/GaAs self-assembled quantum dots to address these questions. While both have been applied separately elsewhere,[8,10] the complementary use of both techniques as a function of electric field provides an important method to probe capture, relaxation, and escape.[11] By using PL excited at differing energies, distinct regimes of carrier escape are identified as a function of field. PC spectroscopy probes the escape directly, and is found to exhibit a very good anticorrelation with the resonantly excited PL. At the low temperatures employed, escape from the dots and wetting layer occurs by tunneling. By varying the field we control the tunneling rate and are able to observe its competition with relaxation and recombination. Good agreement is found with Wentzel–Kramers–Brillouin (WKB) calculations.

The sample studied was a GaAs $p-i-n$ diode structure grown by molecular-beam epitaxy at 500 °C with a single layer of InAs dots embedded at the center of the 0.5 μm GaAs intrinsic region. The dots were formed from 2.4 ML of InAs deposited at 0.39 ML/s using the Stranski–Krastanow technique, and had a density $\sim 5\times 10^{10}\,\mathrm{cm}^{-2}$, heights 35 ± 5 Å, and base lengths 150 ± 10 Å. The electric field F across the intrinsic region is controlled by the built-in potential (V_B) ($\sim$1.5 V) and the applied bias (V_A) according to the relation $F=(V_B-V_A)/w$, where w is the width of the intrinsic region. 400-μm-diam mesas with optical access were employed. PL was excited either by high-energy (1.959 eV, $\sim$1 W cm^{-2}) radiation from a HeNe laser, or by tunable radiation (1.25–1.5 eV, $\sim$300 W cm^{-2}) from a Ti:sapphire laser and dispersed by a 0.22 m monochromator. The PC techniques are described in Ref. 9.

Figure 1 shows quantum-dot ground-state PL spectra as a function of V_A at 10 K for various excitation energies; 1.959 eV in (a) above the GaAs band gap, resonant with the WL transition at 1.445 eV in (b) and directly in the quantum-dot (QD) states at 1.303 eV in (c). In all cases PL from the quantum-dot ground state at 1.14 eV is observed. With above-gap excitation in Fig. 1(a), the PL is strongest under flatband conditions ($F=0$, $V_A=+1.5$ V). As the forward bias is reduced, the intensity of the PL decreases rapidly, fully quenching below $\sim +0.7$ V. In Fig. 1(b) (1.445 eV excitation), the PL intensity changes much less rapidly with bias and is only fully quenched for $V_A<-3$ V. In Fig. 1(c) (1.303 eV excitation) little change is observed between +1 and −3 V, but beyond −3 V, strong quenching is seen, the PL intensity decreasing to zero at −6 V. In addition, the spectra in Fig. 1(c) exhibit a shoulder at 1.21 eV, $\sim$90 meV below the excitation energy, arising from the resonant excitation of dots 3LO phonon energies below the excitation.[12]

The strongly differing behavior in Figs. 1(a)–1(c) can be understood qualitatively by considering the capture and escape processes which occur for the differing excitation energies, as sketched in the diagrams adjacent to Figs. 1(a)–1(c). In Fig. 1(a), carriers must first be captured from the GaAs matrix into the dots in order for dot PL to be observed, whereas in Figs. 1(b) and 1(c), significant WL-dot and intra-dot relaxation only is required. The rapid quenching in Fig. 1(a) occurs before any loss of intensity is in Figs. 1(b) and 1(c), showing that the capture process from the GaAs is very

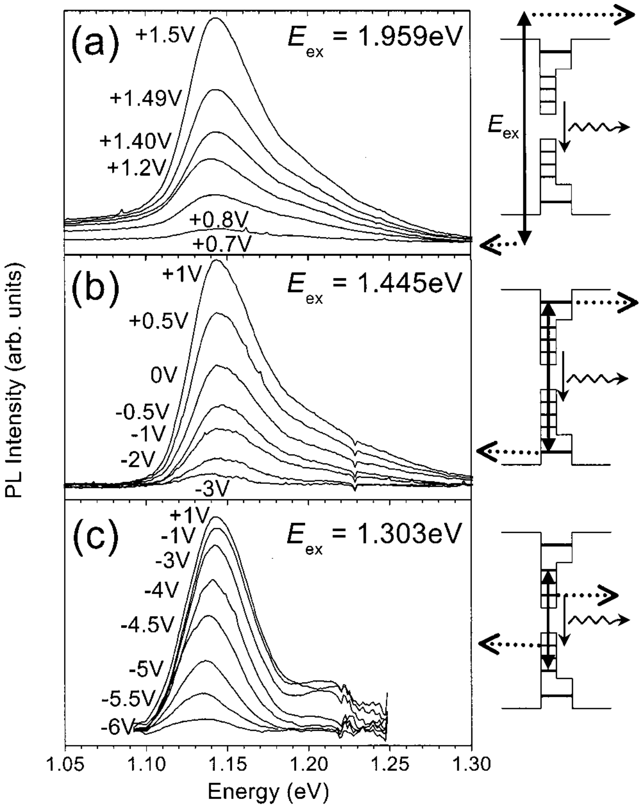

FIG. 1. Photoluminescence spectra at 10 K as a function of bias excited at (a) 1.959 eV above the GaAs band gap, (b) 1.445 eV resonant with the wetting layer, and (c) 1.303 eV resonant with the second dot excited state. Schematic excitation, carrier loss, and recombination processes are indicated for the three cases.

sensitive to the applied electric field. In Fig. 1(b), carriers are excited directly in the WL. Dot PL quenching with field occurs when the tunneling rate from the WL exceeds the relaxation rate from the WL to the dots. Finally, in Fig. 1(c), the PL will be quenched only when the tunneling rate from the dots exceeds the radiative recombination rate.

PC spectroscopy is complementary to PL since it detects those carriers which are excited in the dots and then escape by tunneling [Figs. 1(b) and 1(c), insets], and thus enables the above interpretations to be substantiated [thermal emission is only important at temperatures >150 K (Refs. 7 and 9)]. In the main part of Fig. 2, PC spectra in the spectral region of the dots (~1.1–1.3 eV) are shown. No PC is detected for forward and small reverse bias. As the reverse bias is increased between −2 and −6 V, the spectra develop three features arising from interband transitions of the dots.[9] For higher reverse bias little additional increase in intensity is observed. This behavior demonstrates that carriers can only tunnel out from the dots for $V_A < -2$ V; for $V_A > -2$ V, they relax to the ground state and recombine, [Fig. 1(c)]. PC spectra to higher energy arising from carrier escape from the 2D WL are shown in the inset. For decreasing V_A from +1.5 V, PC at 1.45 eV from the WL is first seen at ~0.5 V, and reaches its full intensity for ~−1.5 V, a lower reverse bias than that required to see *any* QD transitions.

The variations of Figs. 1 and 2 are summarized in Figs. 3(a) and 3(b), where the intensities of the PL and PC are plotted against electric field. In Fig. 3(a), the ground-state PL intensity for excitation into the second excited state is shown

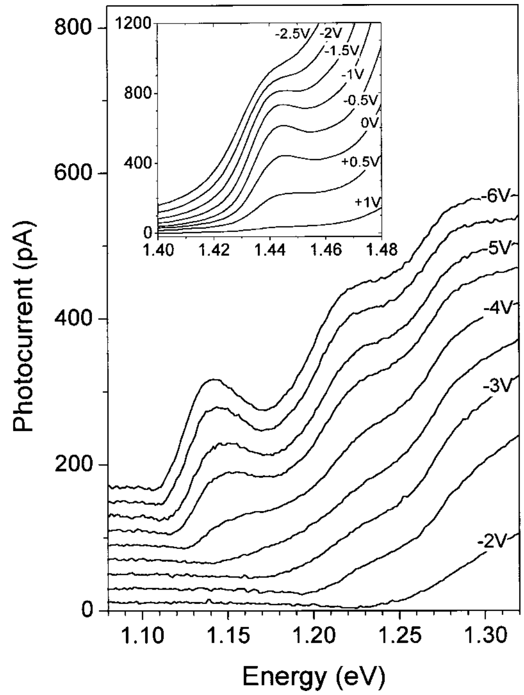

FIG. 2. Photocurrent spectra as a function of bias at 10 K. Quantum-dot features are observed for reverse biases between −3 and −6 V. The inset shows photocurrent from the two-dimensional wetting-layer transition, observed to its full intensity at biases of only ~−0.5 V.

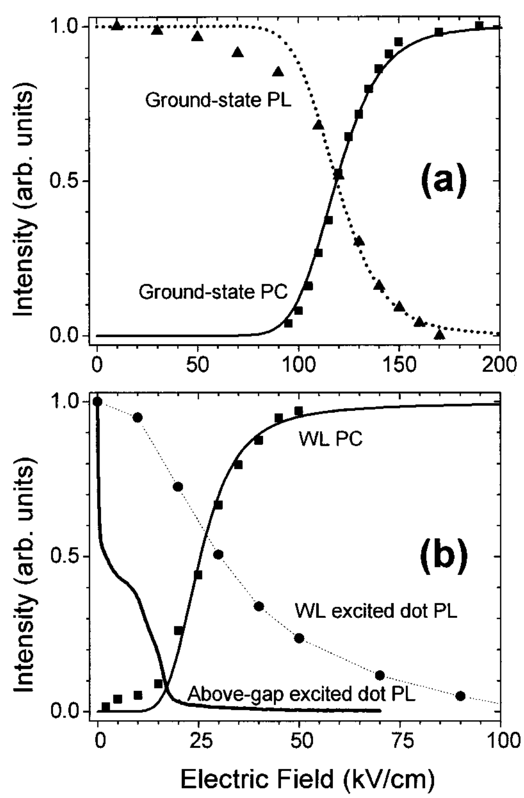

FIG. 3. Summary of photoluminescence (PL) and photocurrent (PC) results of Figs. 1 and 2 plotted as a function of electric field. In (a), the resonantly excited dot PL and dot PC variations are shown. A very good anticorrelation between the two processes is observed, showing that tunnel escape dominates the PL quenching. The full lines are fits to a WKB calculation. In (b), the PL variations with field for above gap and WL excitation are shown, together with the PC from the wetting layer. Very reasonable anticorrelation between the WL PC and the dot PL excited in the WL is found.

(triangles), together with the PC intensity from the ground state (squares). From 0–90 kV/cm the PL decreases by only ~10%, and then between 90 and ~170 kV/cm decreases to zero. The PC intensity shows, to a very good approximation, the inverse dependence, providing strong evidence for competition between tunneling and radiative recombination.

We modeled this behavior using an adiabatic approximation, with decoupled z (parallel to the field) and x,y (in the growth plane) motion. Within this approximation, the tunneling probability depends only on the z part of the wave function, and can be modeled using one-dimensional (1D) WKB calculations.[13] For a 1D confining potential of width L in a perpendicular field F, the tunneling rate R_T is given by

$$R_T=\frac{\hbar\pi}{2m^*L^2}\exp\left[\frac{-4}{3\hbar eF}\sqrt{2m^*E_I^3}\right],$$

where E_I is the ionization energy of the electron eigenstate.[14] At the field (120 kV/cm) where the dot PL is quenched by a factor of 2 and the PC has one half its maximum value, R_T equals the radiative recombination rate ($\sim 1\ \mathrm{ns}^{-1}$).[6] Taking L as 35 ± 5 Å, the dot height, a value for $E_I=190\pm 40$ meV,[15] is deduced from the WKB expression close to the calculated value of 195 meV in Ref. 8 for dots of similar PL energy. Based on this fitting at 120 kV/cm, the expected variation of the ground state PC intensity with field in Fig. 3(a) can be calculated, assuming a Gaussian distribution of states[16] to allow for inhomogeneity. The calculated variation is shown by the full line in Fig. 3(a), and shows excellent agreement with experiment. A reasonably good fit to the ground-state PL data, obtained from the inverse variation of the PC variation, is also obtained (the dashed line).

From Fig. 3(a), it is clear that significant quenching of the (ground-state) PL, excited in the region of the second *excited* state, occurs when the tunneling rate from the *ground* state exceeds the radiative recombination rate.[17] These results show that the intradot relaxation rate (~40 ps) (Ref. 3) is considerably faster than the tunneling rate from the excited states, and that carrier escape occurs predominantly through the ground state.[18] This, in turn, indicates that the excited states have a very similar vertical extent to that of the ground state; if they had more extended character along z, as expected for states arising from z-direction quantization, then tunneling times one to two orders of magnitude less than from the ground state would be expected, and PL quenching at much lower fields would occur. It can thus be further concluded that the excited-state splittings from the ground state arise predominantly from lateral quantization .[9]

Figure 3(b) shows the PL intensity variation with field for 1.959 and 1.445 eV excitation [results of Figs. 1(a) and 1(b)]. For the case of above-gap 1.959 eV excitation, the rapid quenching arises from reduction of the capture rate by the WL and dots from the surrounding matrix. The dot PL intensity for 1.445 eV WL excitation (solid circles) decreases with field to zero over the range 0–100 kV/cm and, to a good approximation, anticorrelates with the WL PC. This good anticorrelation shows that at low fields all carriers excited in the WL relax into the dots and give rise to dot PL. For fields greater than 15 kV/cm, competition between tunnel escape from the WL and WL-QD relaxation occurs with tunneling dominant above ~40 kV/cm. In this case, tunnel escape competes with WL-dot relaxation occurring on an ~10 ps time scale,[6] as opposed to radiative recombination in the case of the resonant excitation above (1 ns time scale). Equating R_T to 10 ps at 27 kV/cm, and assuming $L=20$ Å and linewidth 20 meV, an E_I value of 45 meV, very reasonable for the WL energy of 1.445 eV, was obtained and led to the good fit to the PC with field in Fig. 3(b).

In summary, we have identified differing regimes of electric-field quenching of photoluminescence from InAs quantum dots. For above-gap excitation, inhibition of capture is found at very small electric fields (<10 kV/cm). For carriers excited in the wetting layer, direct capture into the dots dominates at low field (<15 kV/cm), but with increasing field tunnel escape from the WL dominates (>40 kV/cm). For direct excitation in the dots, the occupation of the active region is much more stable, with tunnel escape only important at fields in excess of 100 kV/cm. These conclusions are strongly supported by photocurrent measurements.

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[1] D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, New York, 1998).
[2] J. J. Finley, M. Skalitz, M. Arzberger, A. Zrenner, G. Böhm, and G. Abstreiter, Appl. Phys. Lett. **73**, 2618 (1998).
[3] R. Heitz, M. Veit, N. N. Ledenstov, A. Hoffmann, D. Bimberg, V. M. Ustinov, P. S. Kop'ev, and Zh. I. Alferov, Phys. Rev. B **56**, 10435 (1997).
[4] A. V. Uskov, J. McInerny, F. Adler, H. Sweizer, and M. H. Pilkuhn, Appl. Phys. Lett. **72**, 58 (1998).
[5] R. Ferreira and G. Bastard, Appl. Phys. Lett. **74**, 2818 (1999).
[6] B. Ohnesorge, M. Albrecht, J. Oshinawa, A. Forchel, and Y. Arakawa, Phys. Rev. B **54**, 11532 (1996).
[7] C. M. A. Kapteyn, M. Lion, R. Heitz, D. Bimberg, P. N. Brounkov, B. V. Volovik, S. G. Konnikov, A. R. Kovsh, and V. M. Ustinov, Appl. Phys. Lett. **76**, 1573 (2000).
[8] C. M. A. Kapteyn, P. Heinrichsdorff, O. Stier, R. Heitz, M. Grundmann, N. D. Zakharov, D. Bimberg, and P. Werner, Phys. Rev. B **60**, 14265 (1999).
[9] P. W. Fry *et al.*, Phys. Rev. Lett. **84**, 733 (2000).
[10] K. H. Schmidt, U. Kunze, G. Medeiros-Ribeiro, J. M. Garcia, P. Wellmann, and P. M. Petroff, Physica E (Amsterdam) **2**, 627 (1998).
[11] S. Tarucha, K. Ploog, and K. von Klitzing, Phys. Rev. B **36**, 4558 (1987).
[12] M. J. Steer, D. J. Mowbray, W. R. Tribe, M. S. Skolnick, M. D. Sturge, M. Hopkinson, A. G. Cullis, C. R. Whitehouse, and R. Murray, Phys. Rev. B **54**, 17738 (1996).
[13] D. M.-T. Kuo and Y. C. Chang, Phys. Rev. B **61**, 11051 (2000); see reference therein to three-dimensional tunneling rate calculations for QDs; the results agree within a factor of 3 with those obtained from 1D quantum-well calculations, justifying the approach adopted here.
[14] The electron tunneling rate is 5–10 orders of magnitude faster than that of holes due to the 5–7 times smaller electron mass, see, e.g., Ref. 2. Tunneling is thus initiated by electrons and, therefore, employed in the calculations. Hole space charge builds up until the electron and hole fluxes out of the dots are equal. The space-charge buildup is, nevertheless, small and does not effect the electrostatics, as verified by power-dependent measurements.
[15] The error is due mainly to uncertainties of the electron effective mass over the range $0.05-0.067m_e$.
[16] A Gaussian centered at 190 meV is used width of 25 meV, approximately half the linewidth of the PC/PL. We assume the broadening is split equally between electron and hole fluctuations.
[17] Resonant PL was performed at several other excitation energies (1.22–1.30 eV) and while the spectra became increasingly dominated by LO-phonon replicas as the excitation energy was reduced (see Ref. 12), the field dependence of the ground-state PL intensity remained the same.
[18] The observation of all the interband PC transitions at the same bias also shows that escape occurs through a common path (the ground state).