Electronic energy levels and energy relaxation mechanisms in self-organized InAs/GaAs quantum dots

M. J. Steer,* D. J. Mowbray,† W. R. Tribe,‡ M. S. Skolnick, and M. D. Sturge§
Department of Physics, University of Sheffield, Sheffield S3 7RH, United Kingdom

M. Hopkinson, A. G. Cullis, and C. R. Whitehouse
Engineering and Physical Sciences Research Council Central Facility for III-V Semiconductors, Department of Electronic and Electrical Engineering, University of Sheffield, Sheffield, S1 3JD, United Kingdom

R. Murray
Department of Material Science and Engineering, University of Liverpool, Liverpool, L69 3BX, United Kingdom
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We report a spectroscopic investigation of the electronic energy levels and carrier-relaxation mechanisms in self-organized InAs/GaAs quantum dots. Power-dependent photoluminescence (PL) and photoluminescence excitation (PLE) are used to study the energy-level structure. Two excited states, 74 and 120 meV above the luminescent ground state, are identified. As expected for a zero-dimensional system, it is not possible to observe PL from the ground state of the dots when exciting between the energies of the ground and first excited state due to the discrete, atomiclike nature of the electronic states. Selectively excited PL and PLE reveal two mechanisms for the relaxation of carriers from the excited states to the ground state: a nonresonant mechanism dominant in the upper state, and a resonant mechanism, involving the emission of one or more LO phonons of well-defined energy, which is dominant in the lower excited state. The resonant mechanism is shown to be a consequence of the distribution of energy-level spacings in the inhomogeneous ensemble of dots; preferentially selecting dots with an energy-level spacing close to an integer multiple of the LO phonon energy. [S0163-1829(96)02548-9]

I. INTRODUCTION

Quantum dots provide the ultimate quantum system with three-dimensional carrier confinement resulting in atomiclike, discrete electronic energy states. In addition to allowing the study of physics in a zero-dimensional semiconductor system, these discrete energy levels are expected to result in a number of advantageous properties for electronic and electro-optic device applications. Quantum dot lasers are predicted to exhibit both low threshold current densities,[1] and low- or zero-temperature dependence of the threshold current,[1] while the use of quantum dots may offer possibilities for low-power nonlinear devices. However, for device applications to be a realistic prospect the quantum dots must satisfy a number of requirements. These include large carrier confinement and energy-level separations $\gg kT$, large areal densities, high optical quality, and uniform size and shape.

Of the many techniques proposed and investigated for the fabrication of quantum dots, perhaps the most promising is that of self-organized growth.[2–5] Dots prepared by this technique appear to be capable of satisfying all of the above requirements, although further improvements in size and shape uniformity are desirable. Self-organized growth may occur when a thin layer of one semiconductor is grown epitaxially on a second semiconductor of a different lattice constant. For intermediate values of lattice mismatch the initial two-dimensional growth transforms, above a certain critical thickness, to nonuniform three-dimensional growth, resulting in a spatial modulation of the epitaxial layer thickness. This is known as the Stranski-Krastanov growth mechanism. The small areas of three-dimensional growth, which sit on a thin two-dimensional layer (the so-called wetting layer), form the quantum dots. Although initially observed in the InAs-on-GaAs system, self-organized dots have now been observed in a wide range of material systems.[4,6,7] For the InAs-on-GaAs system, for which there is a 7% lattice mismatch, the resultant InAs dots have a typical base size $\sim$10–25 nm and height $\sim$2–10 nm,[2,8] the actual size being dependent to some extent upon the growth conditions. These dimensions are small enough that strong quantum effects are observed.

In this paper we present a study of the electronic energy levels and carrier relaxation mechanisms in self-organized InAs/GaAs quantum dots. The latter topic is of particular importance in zero-dimensional systems since it has been predicted that their discrete, atomiclike energy levels may inhibit the efficient carrier relaxation by single phonon emission, which occurs in systems with continuous energy levels.[9] Unless other efficient relaxation mechanisms are possible, i.e., multiphonon,[10] Auger,[11] or long-range resonance energy transfer,[12] carrier relaxation rates will be very slow, with serious implications for device performance.

II. EXPERIMENTAL DETAILS

The samples were grown by solid source molecular-beam epitaxy using conditions very similar to those of Moison *et al.*[2] The structure consisted of a thin layer of InAs deposited on a GaAs buffer layer, which in turn was grown on an undoped GaAs substrate. At the growth temperature used (T_g=500–520 °C) the transformation from two- to three-

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dimensional growth was found to occur after the deposition of a nominal 1.8–1.9 ML of InAs. However, x-ray analysis of samples containing an $In_xGa_{1-x}As/GaAs$ multiple-quantum-well buffer suggests that the InAs thickness may be ~5–10 % lower than intended due to In desorption at the growth temperature used. The main sample studied in this paper consisted of 2.4 ML (nominal) of InAs, which resulted in square quantum dots of base length ≈12 nm, height ≈2 nm, and density $\sim 5\times10^{10}$ cm^{-2}, as evidenced by plan view and cross-sectional transmission electron microscopy.[13] The growth was terminated with the InAs dots being overgrown by a GaAs capping layer of thickness 100 nm.

Photoluminescence (PL) and high-resolution photoluminescence excitation (PLE) spectra were excited with an Ar^+ pumped Ti: sapphire laser and the resultant PL was dispersed by a double-grating spectrometer and detected with a liquid-nitrogen-cooled Ge *p-i-n* photodiode. Low-resolution PLE spectra were obtained over a wider energy range and at lower incident power density with a 150-W tungsten halogen lamp and monochromator combination. In both cases a large number of dots (~10^8 dots for laser excitation with a cylindrical focusing lens illuminating an area $\approx 2\times10^{-3}$ cm^2 and ~10^{10} dots for lamp and monochromator excitation of an area ≈0.1 cm^2) were probed. Higher incident laser power densities ~50–5000 W cm^{-2} were obtained with a micro-PL system that uses a HeCd laser (λ_{ex}=440 nm excitation) and a high-quality microscope objective to give a laser spot size ~1–2 μm. Most of this short-wavelength excitation is absorbed in the GaAs capping layer resulting in strong excitation of the dots (~1 *e-h* pair per dot at 50 W cm^{-2}). All the measurements were performed with a sample temperature of 4.2 K.

III. EXPERIMENTAL RESULTS

The quantum dot samples exhibit very strong PL at ≈1.1 μm, for excitation both above and below the GaAs barrier band gap. We compared their PL efficiency to that of high-quality 10- and 20-Å $Ga_xIn_{1-x}As$-InP quantum wells, which emit in the same wavelength region and are believed to have very high luminescent efficiency.[14] For both types of structure the excitation was directly into the active region using photons of an energy less than that of the barrier band gap. The integrated PL intensity of the dot sample was found to be a factor ~10 weaker than that of the individual quantum wells. However, given that the dots occupy only ~5% of the total sample area, the absolute quantum efficiencies of the dots and the quantum wells are comparable. This result suggests that the dots do not introduce a significant number of nonradiative recombination pathways.

Figure 1 shows PL spectra as a function of incident laser power, using the high-power densities obtainable with the micro-PL system. If we assume a radiative lifetime for the dots of ~1 ns,[15] no lateral carrier diffusion within the wetting layer away from the excited region and that all the photoexcited carriers produced in the GaAs barriers are subsequently captured by the dots, the lowest power of 45 W cm^{-2} is calculated to produce a dot occupancy ~1. At this power we observe only a single peak at 1.13 eV, labeled T_1. We will refer to this as the ''ground-state'' transition. The width of this peak (35 meV at the lowest power) results from variations in the size and shape, and hence variation in the ground-state energy, of the dots probed. This linewidth is somewhat smaller than values previously reported for similar samples,[3,5,16] indicating that these size and shape fluctuations are reasonably small [a 35-meV PL linewidth corresponds to base length variations of ≈±0.5 nm (Ref. 17)]. With increasing laser power density the intensity of the ground-state emission approximately saturates and additional features T_2 and T_3 are observed to higher energies. This behavior is attributed to the filling of the ground state and the resultant recombination from excited states.[18,19]

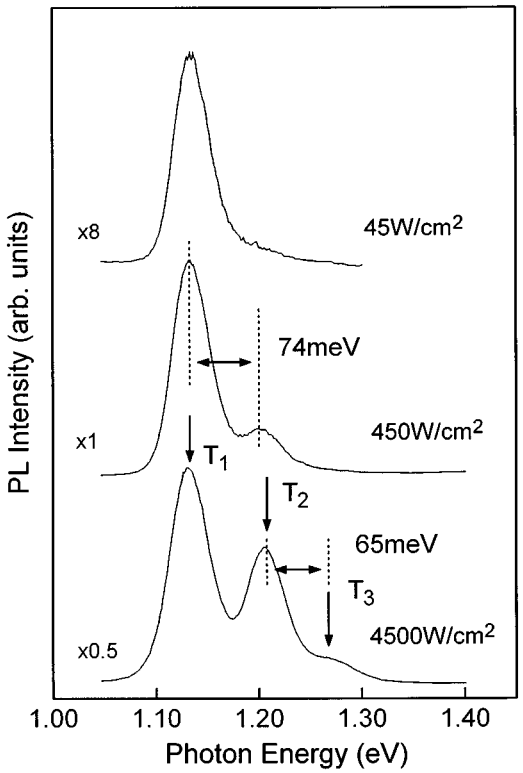

FIG. 1. Power-dependent PL spectra of the quantum dot sample excited with short-wavelength light (λ_{ex}=4420 Å≡2.80 eV).

The nature of the excited dot states is unclear with calculations of the electronic structure of self-organized InAs dots giving conflicting results. For example, the calculations of Grundmann, Stier, and Bimberg[17] for pyramidal dots predict only one confined electron state, with allowed transitions to a number of heavy-hole states. On the other hand, Marzin and Bastard[20] predict at least two confined electron states for cone-shaped dots. A possible cause of this discrepancy is the different choice of effective mass for the electrons. Capacitance-voltage measurements, which distinguish electron from hole states, by Medeiros-Ribeiro, Leonard, and Petroff[21] on ≈20-nm-diameter InAs dots indicate two electron states but only one hole state. In view of this uncertainty concerning the electronic structure of the dots we cannot reliably identify the states involved with the transitions T_2 and T_3. The conclusions reached in the remainder of this paper are independent of such identification.

Although PLE has been used extensively to study the excited states of quantum wells, the observed PLE spectrum only represents the absorption spectrum if the probability of relaxation from the initially excited state to the ground (luminescing) state is independent of energy. Any energy de-

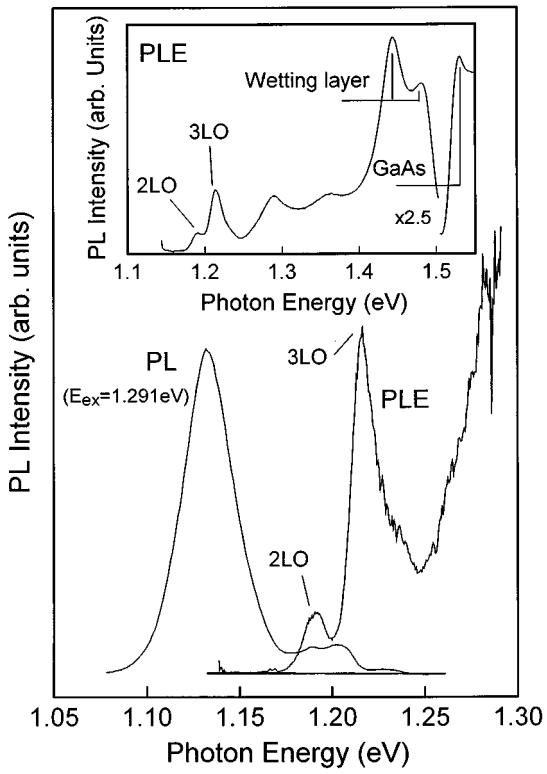

FIG. 2. PL and PLE spectra obtained using laser excitation. The inset shows a typical PLE spectrum recorded using lamp and monochromator excitation. For both PLE spectra detection is at the PL maximum.

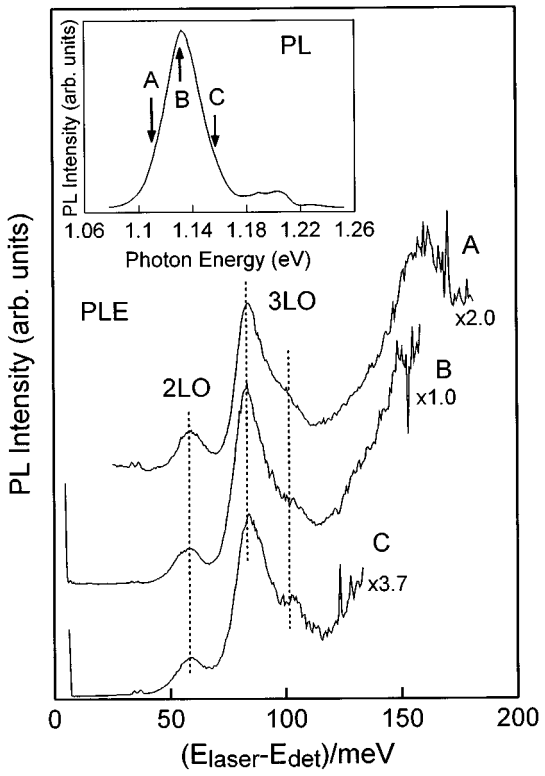

FIG. 3. PLE spectra obtained for different detection energies as indicated in the inset. The spectra are plotted as a function of the energy difference between the incident photon energy and the detection energy.

pendence of this probability will distort the spectrum. In quantum wells this is generally a weak effect and PLE provides reliable excited-state transition energies. However, we find that in our quantum dots the carrier relaxation efficiency is a function of energy, showing strong resonances when the energy separation between the incident photon and detection energies is equal to certain integer multiples of the LO phonon energy. This energy dependence dominates the spectra, so that excited-state energies cannot be determined directly from the spectra. This behavior is shown in the main part of Fig. 2, which shows PL (excited at 1.291 eV) and PLE (detected at 1.14 eV, the peak of the PL) of the sample, both recorded with laser excitation. The absolute energies of the sharp features in the PLE spectrum, labeled 2LO and 3LO, depend upon the detection energy, shifting rigidly as this energy is varied (see Fig. 3 below). Hence the peak positions do not give the excited-state energies, and we will show below that they appear to represent the effects of carrier relaxation by the emission of two or more LO phonons of well-defined energy.

For energies below the phonon features ($\lesssim$1.17 eV) the PLE signal falls to zero. This indicates that no measurable PL is produced when exciting in this energy range, which corresponds to energies between the ground state and first excited state of the dots being probed. Such behavior is consistent with that expected for a zero-dimensional system with discrete, atomiclike energy levels. Although light may be absorbed directly into the ground state of the dots and will subsequently be reemitted, for a zero-dimensional system this should occur at the same energy as that of the incident photons (zero Stokes shift), so long as the dots are sufficiently far apart that no transfer between dots occurs.[22] Such resonant ground-state emission has not been observed using our present experimental techniques, probably because it is obscured by the elastically scattered laser light from the sample surface or by resonant Rayleigh scattering from the inhomogeneously broadened ground-state transition.[23] Hence, for the present experimental conditions, only by absorbing into an excited state, from which relaxation occurs to the lower-energy emitting ground state, can a nonzero PLE signal be obtained.

The inset to Fig. 2 shows a PLE spectrum recorded using the lamp and monochromator combination, which allows a much wider energy range to be scanned than is available with the laser. For low energies the spectrum agrees with that obtained with the laser. At higher energies the lamp and monochromator PLE spectrum shows absorption occurring into both the two-dimensional InAs wetting layer ($\approx$1.4–1.5 eV) and the GaAs barrier layers ($\approx$1.52 eV). The features occurring at $\approx$1.29 and $\approx$1.37 eV are briefly discussed below.

The behavior of the features labeled 2LO and 3LO in the PLE spectrum of Fig. 2 is shown in more detail in Fig. 3, which shows PLE spectra plotted against the difference between the incident photon energy and the detection energy. Spectra are shown for detection at three different points in the PL peak shown in the inset (recorded for an excitation energy of 1.291 eV). The PLE spectra show three peaks with constant shifts from the detection energy. For the present sample these energy shifts are 58(2×29), 84(3×28), and

102(3×34) meV, respectively. The unstrained LO phonon energy of InAs is 29.9 meV and the calculations of Grundmann, Stier, and Bimberg[17] suggest that strain increases this value to 32.1 meV in pyramidal-shaped dots. In the wetting layer a slight reduction to 29.6 meV is predicted, due to the competing effects of strain and confinement.[17] Hence the features observed in the PL and PLE spectra occur at approximately two (2LO) and three (3LO) multiples of these InAs LO phonon energies. We therefore tentatively identify them as being due to carrier relaxation processes involving the emission of two or three LO phonons. Although possibly two 3LO features (84 and 102 meV) are present in the spectra of Fig. 3, which may indicate the coupling of the dot electronic states to both the dot and wetting layer LO phonons, only one 2LO feature is observed. However, it is possible that the 2LO feature corresponding to the weak 3LO feature at 102 meV is obscured by the low-energy tail of the dominant 3LO feature at 84 meV.

The present results are analogous to those of Fafard *et al.*,[18] where strong features shifted by a single LO phonon from the excitation energy were observed in the PL spectra of $Al_xIn_{1-x}As/Al_xGa_{1-x}As$ self-organized quantum dots, and those of Heitz *et al.*[16] for InAs/GaAs dots similar to ours. However, the sample of Ref. 16 had a slightly larger PL linewidth (45 meV),[16] and a peak was observed in PLE at the single (1LO) phonon energy in addition to the 2LO and 3LO features observed in the present sample. As will be explained later, the phonon features are believed to arise because the experiment selects out of the ensemble of dots having a range of energy-level separations,[16] those dots for which this separation is an integral number of LO phonon energies. In our relatively homogeneous samples, the smallest energy separation is presumably greater than the single LO phonon energy.

Heitz *et al.*[16] also observed substructure in their multiple phonon peaks, as measured in resonantly excited PL, which they attributed to coupling to various different phonon modes including interface, dot, wetting layer, and GaAs phonons. Such substructure is not observed in the present sample. The very sharp lines observed at ~35 meV are Raman lines. These have energies of 33.7 and 36.5 meV, corresponding to the bulk GaAs TO and LO phonon energies, respectively, and do not show significant resonance effects in the energy region of the dot states. They thus represent bulk GaAs phonons that are not coupled to the dot electronic states.

Figure 4 shows a series of PL spectra recorded for a range of excitation energies. For high excitation energies the spectra are dominated by a broad peak (indicated by the dashed line, which is a Gaussian fit to the data) having a roughly constant energy of ~1.125 eV, which, by comparison with the spectra of Fig. 1, is attributed to the ground-state emission from the quantum dot ensemble. We refer to this broad feature as the ''nonresonant ground state'' (NRGS) emission. With decreasing excitation energy the intensity of the NRGS emission decreases and a number of sharper features are observed, the absolute energies of which depend upon the laser excitation energy. The approximately constant energy separation of these features from the laser energy allows them to be identified as the PL equivalents of the 2LO and 3LO features observed in the PLE spectra of Figs. 2 and 3. Because the intensities of the phonon features increase as their emission energies approach the maximum of the nonresonatly excited PL, their relative intensities in individual PL spectra are different from those observed in the PLE spectra of Fig. 3. For example, for excitation energies ≈1.245 eV the normally weak 3LO feature at 102 meV (see Fig. 3) is close to resonance and is hence relatively intense. However, its intensity decreases rapidly as it moves off resonance with decreasing laser excitation energy.

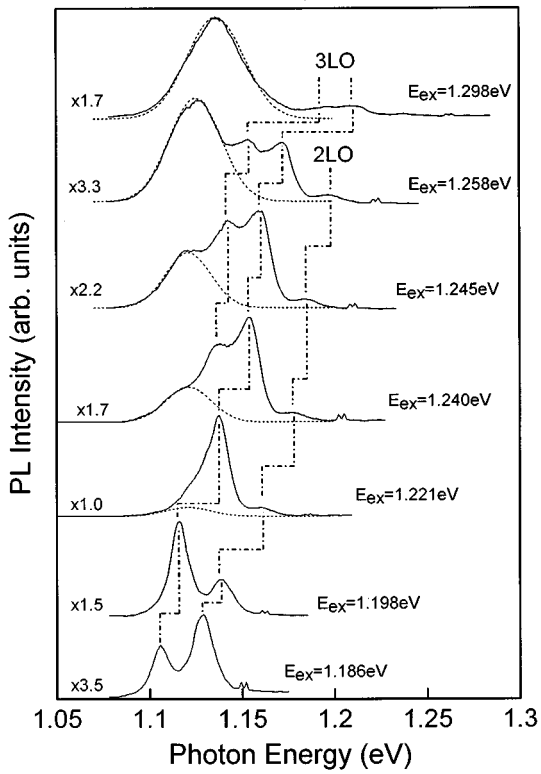

FIG. 4. Selectively excited PL spectra obtained for a range of excitation energies. The dashed lines indicate a Gaussian fit to the nonresonant ground state (NRGS) emission. The dot-dashed lines indicate how the resonant features follow the excitation energy (note that as in PLE there are two features assigned to 3LO).

In the main part of Fig. 5 the intensities of the NRGS and the 2LO and 3LO phonon features, as observed in PL, are plotted against the incident photon energy. A typical PL spectrum obtained with excitation at 1.959 eV, above the GaAs band gap, is also shown for comparison. In the inset the intensities of the phonon features are plotted against their respective emission energies. Except for intensity the phonon curves plotted in the inset are essentially identical to the PL obtained with above GaAs band-gap excitation, showing that the same distribution of luminescent states is probed by each process. In particular, this indicates that the strengths of the individual 2LO and 3LO multiple-phonon resonant relaxation features are determined by the density of final (ground) radiative states.

The PL spectra of Fig. 4 and the data plotted in the main part of Fig. 5 suggest that carriers can relax to the ground state of the dots by two distinct mechanisms, which we refer to as ''nonresonant'' and ''resonant,'' respectively. The relative importance of these two mechanisms depends upon the excitation energy. For high-energy excitation (≳1.23 eV, the

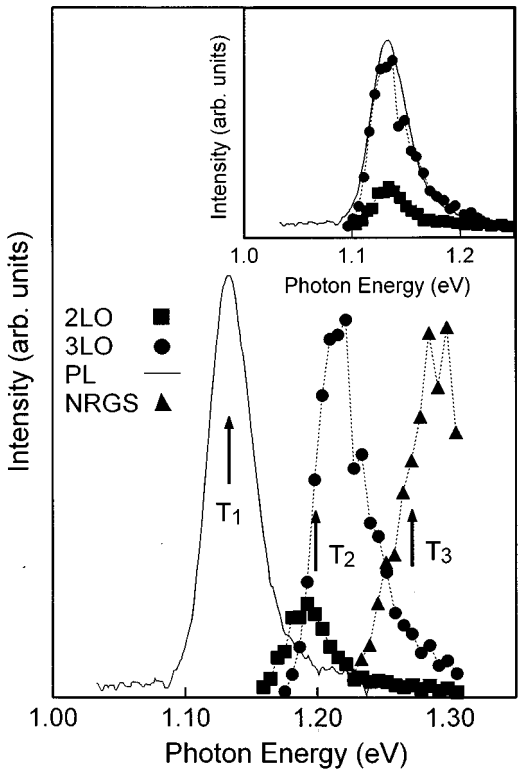

FIG. 5. Intensities of the various components of the ground-state emission as observed in resonantly excited PL. In the main figure the intensities of the NRGS 2LO, and 3LO emissions, obtained from Fig. 4, are plotted against the excitation energy. In the inset they are plotted against the relevant emission energy. Only the more intense 3LO phonon feature, of energy 84 meV, is plotted. Also shown is a typical PL spectrum obtained with nonselective excitation above the GaAs band gap (λ_{ex}=6328 Å≡1.959 eV). The positions of the optical transitions, as determined from high incident power PL spectra, are indicated by the vertical arrows.

nonresonant NRGS excitation regime) broad emission is observed, indicating that the ground states of a significant fraction of the dot ensemble are being populated. The PL peak shifts downwards by about 14 meV as the excitation energy is reduced by 53 meV from 1.298 eV (see Fig. 4) showing that the excitation is somewhat selective due to the correlation between the excited- and ground-state energies of the different-sized dots.[7] The ratio 53:14 is consistent with the calculated dependence of the ground- and excited-state energies on dot size.[16,17] However, although the width of the PL peak varies slightly, decreasing from 36 to 28 meV as the excitation energy is reduced from 1.298 to 1.245 eV, it remains fairly broad, indicating that the relaxation mechanism responsible for this emission is nonresonant in nature. Possible nonresonant mechanisms for relaxation in this regime include the emission of continuum (nonzero k) phonons, cascade through nonradiating excitonic states[24] or tunneling between dots and subsequent resonant or nonresonant relaxation. While this broad nonresonant PL ceases to be observable for excitation below 1.22 eV, it may still be present, but obscured by the stronger resonant emission. However, its intensity for excitation below 1.22 eV does not exceed 15% of the intensity for 1.3-eV excitation.

With decreasing excitation energy below ~1.30 eV the intensity of the NRGS decreases. However, relaxation to the ground state now becomes possible by a resonant process involving the emission of either two or three LO phonons. These processes become important when the energy difference between the incident photon and the dot ground state is close to a multiple of the LO phonon energy ($2\hbar\omega_{LO}$ or $3\hbar\omega_{LO}$). The relatively narrow linewidths (~10 meV) of the phonon features suggest that, unlike the first process, relaxation to only a small fraction of the dot ground states is possible for a given excitation energy. The linewidth is probably nonzero for two reasons: a low-energy LA phonon can be involved in addition to the LO phonon,[25] and the LO phonon itself can have a spread in energy due to strain.[17] The experimental data therefore suggest the presence of two carrier relaxation mechanisms. The first is nonresonant and permits relaxation to a large fraction of the ground states of the dot ensemble, any selectivity being due solely to the electronic structure. The second, resonant mechanism, involves the emission of two or three LO phonons of well-defined energy and populates the ground states of only a small fraction of the dot ensemble.

The energies of the dot transitions, as determined from the high incident laser power intensity PL spectra of Fig. 1, are indicated in Fig. 5. It appears that the nonresonant mechanism is associated with absorption at the second excited-state transition T_3, and is weaker by a factor of at least 6 for the lower-energy transition T_2. The resonant process, on the other hand, involves initial absorption at the T_2 transition followed by resonant relaxation to the ground state by multiple LO phonon emission. The relaxation is primarily through the emission of three LO phonons at higher excitation energies and two at lower energies. The reason for the contrasting dependence of the two relaxation mechanisms on excitation energy is not fully clear. However, a possible explanation is that the nonresonant process involves interdot tunneling since this should be stronger for the higher-energy state, where the height of the tunneling barrier is smaller, thus favoring the involvement of the T_3 states in the NRGS emission. A nonresonant tunneling process is required, possibly acoustic phonon assisted as has been observed between the ground states of very closely spaced quantum dots.[22] The relaxation mechanism would hence consist of the initial nonresonant tunneling between dots followed, after on average at least one tunneling event, by intradot relaxation, possibly by the emission of multiple LO phonons. Approximately similar rates for the interdot tunneling and the intradot energy relaxation are required to ensure an overall quasinonresonant process. For example, a faster tunneling rate would allow carriers to reach the largest dots before relaxation to the ground state occurred whereas a faster intradot relaxation rate would result in the resonant relaxation observed for absorption into the lower-energy T_2 state. However, we stress that until the form of the nonresonant relaxation process is unambiguously known, a detailed understanding of the mechanisms involved is not possible. It is also important to note that other explanations for the nonresonant relaxation mechanism exist. For example, the upper excited level could consist of a number of closely spaced, unresolved states between which initial acoustic phonon assisted intradot relaxation occurs followed by multiple LO phonon relaxation to the ground state.

An explanation for the selectivity of the resonant process has been proposed by Heitz *et al.*[16] Unlike quantum wells where there is only one variable dimension (the well width), dots having different combinations of base length and height may have the same ground-state energy but different excited-state energies. As different ground-state energies are probed by varying the PLE detection energy, those dots having a level spacing equal to an integer multiple of the LO phonon energy are the only ones that can relax to the ground state by LO phonon emission. Hence the PLE spectra should show a constant shift from the detection energy, as observed experimentally, the shift being determined by the phonon energies rather than by the electronic level splitting. A further consequence of this mechanism is that carriers created in dots having energy-level spacings not resonant with an integer number of LO phonons are lost from the dot before they can relax to the ground state. It has been suggested that such carriers could transfer to deep levels in the GaAs barrier layers.[16] However, a more likely explanation is that these carriers recombine radiatively from the excited states, the resultant PL occurring at the excitation energy and consequently not being observable using the present experimental technique.

Calculations suggest that the range of excited-state energies for a given ground state, resulting from dot size and shape fluctuations, is sufficient in the sample of Heitz *et al.*[16] to account for their observation of constant energy multiple phonon related features. Our data confirm this. We have studied a number of other samples with differing degrees of dot size and shape inhomogeneity, as evidenced by the PL linewidth. Multiple phonon related carrier relaxation features are observed in all samples, the number of such features scaling approximately with the degree of inhomogeneity (i.e., with the width of the distribution of energy-level spacings). Figure 6 compares two samples with different PL linewidths, and hence different degrees of dot homogeneity, with results consistent with the interpretation given above. Fig. 6(b) shows PLE and resonantly excited PL spectra of the sample discussed extensively above, while Fig. 6(a) shows the corresponding spectra of a second sample, grown under similar conditions, whose larger nonresonant PL linewidth of 67 meV indicates a greater spread of dot sizes. While the PL of the sample with linewidth 35 meV shows only 2LO and 3LO features, the sample with the larger PL linewidth shows, in addition, 1LO, 4LO, and 5LO features. A correspondingly greater number of phonon features is observed in the PLE spectrum of the more inhomogeneous sample. In addition, the PLE spectrum of the more homogeneous sample [Fig. 6(b) and the inset to Fig. 2] shows two features at higher energies (≈160 and 230 meV) the absolute energies of which also shift rigidly with the detection energy. However, corresponding features at similar energies are not observed in suitably resonantly excited PL spectra and the origin of these features is at present not understood.

The two samples show some difference in the energy of the fundamental phonon mode as measured in PLE. For the more homogeneous sample [Fig. 6(b)] features occur at 58(2×29) and 84(3×28) meV for 2LO and 3LO, respectively, whereas the features in the less homogeneous sample [Fig. 6(a)] occur at 32.3, 67.6(2×33.8), 97(3×32.3), 128(4×32), and 168(5×33.6) meV for 1LO, 2LO, 3LO, 4LO, and 5LO, respectively. It is not clear if this difference represents coupling to different phonons in the two samples or coupling to an identical phonon, the energy of which is sample dependent.

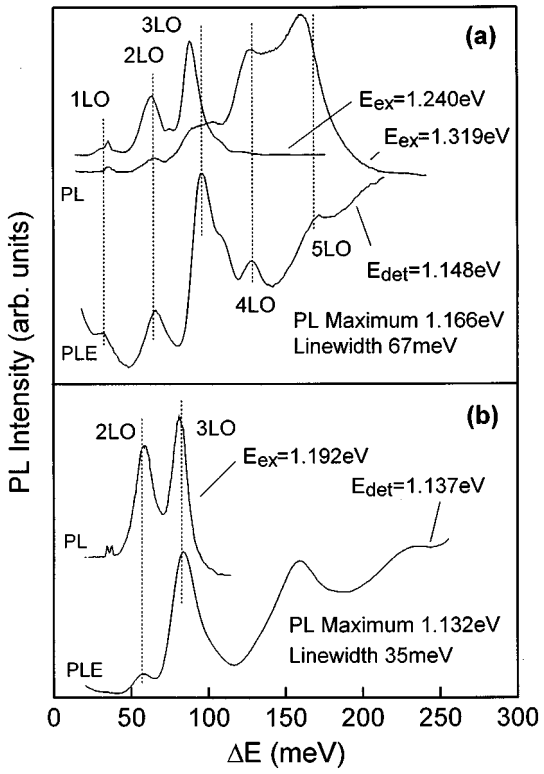

FIG. 6. Resonantly excited PL and PLE spectra for (a) a sample with a ground-state PL linewidth of 67 meV indicating a large dot size and shape inhomogeneity, and (b) the more homogeneous sample studied extensively in this paper having a PL linewidth of 35 meV. The selectively excited PL spectra are plotted against the difference between the incident laser energy and that of the emitted photons ($\Delta E = E_{ex} - E_{photon}$) and are hence reversed compared to those plotted in previous figures. The PLE spectra are plotted against the difference between the incident photon energy and the detection energy ($\Delta E = E_{photon} - E_{det}$).

IV. CONCLUSIONS

We have presented a study of the electronic energy levels and carrier relaxation mechanisms of self-organized InAs/GaAs quantum dots of base length ≈12 nm and height ≈2 nm. Power-dependent PL shows that there are at least three optically allowed transitions, at 1.136, 1.210, and 1.275 eV (averaged over the dot size distribution), respectively. The relatively narrow linewidth of the ground-state emission (35 meV in our best sample) indicates a small quantum dot size distribution. PLE and resonantly excited PL spectra show evidence for two different carrier relaxation mechanisms, a nonresonant process, and a resonant process involving the emission of one or more LO phonons of well-defined energy, independent of dot size. These mechanisms bypass the phonon relaxation bottleneck predicted for zero-

dimensional systems. They differ in their dependence on the excitation energy, the former being dominant in relaxation from the higher (1.275 eV) excited state, the latter from the lower (1.210 eV) state. By studying samples exhibiting different degrees of dot size and shape inhomogeneities we obtain evidence supporting the recent proposal[16] that the resonant relaxation mechanism is a consequence of the distribution of energy-level separations in an inhomogeneous ensemble of dots.

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[*]Also at Department of Electronic and Electrical Engineering, University of Sheffield, Sheffield, S1 3JD, UK.

[†]Electronic address: D.Mowbray@Sheffield.ac.uk

[‡]Present address: Cavendish Laboratory, Department of Physics, University of Cambridge, Cambridge, CB3 OHE, UK.

[§]Permanent address: Physics Department, Dartmouth College, Hanover, NH 03755-3528.

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