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Tuning InAs/GaAs Self-Assembled Quantum Dots to investigate relaxation processes

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ABSTRACT

Using a combination of resonant and non-resonant photoluminescence and photoluminescence excitation spectroscopy we have investigated the carrier relaxation mechanisms in annealed InAs/GaAs self-assembled quantum dots. We find no evidence for LO phonon resonances in the PLE spectra and the data are consistent with absorption into the excited states of the dots. For samples annealed at the highest temperatures the excitation spectra exhibit many features, which we attribute to absorption by "forbidden" transitions involving confined hole states.

INTRODUCTION

There is as yet no consensus regarding the relaxation mechanisms that operate in self-assembled quantum dots (QDs). Indeed it is not known whether the carriers relax as excitons or independent electrons and holes. Fast relaxation on a sub-picosecond scale through the generation of LO phonons is known to dominate in higher dimensional systems but in principle this mechanism would be inhibited in ideal QD systems where a "phonon bottleneck" [1] would result if the separation of the electronic states was not equal to an LO phonon (or multiple LO phonon). Although it is generally accepted that several LO phonons may be available in this system[2] this would still place significant restrictions on the range of inter-level separations which can participate in fast relaxation. However, many studies have demonstrated strong emission from QDs, casting doubt on the phonon bottleneck process. Acoustic phonons can also participate in the relaxation processes although at a significantly smaller rate than those of LO phonons. Nevertheless multi-phonon scattering involving acoustic and LO phonons is predicted to lead to relaxation lifetimes of the order of a few tens of ps in good agreement with time resolved studies[2-5]. It has even been suggested that phonon assisted emission during resonant photoluminescence (PL) is not due to real crystal states but rather is a manifestation of the Fröhlich interaction between strain-induced polarized excitons and LO phonons[6]. Phonons, however, are not the only possible relaxation mechanism and it has been pointed out that the presence of carriers in the dots and wetting layer can lead to efficient Auger and carrier-carrier scattering[7,8,9] and this may well dominate the relaxation rates, particularly under high excitation. Finally, Sercel[10] has proposed a novel mechanism involving transfer of excess carrier energy to grown-in point defects close to the dots. However, recent work on injecting hydrogen into QDs[11], which would be expected to passivate these beneficial grown-in defects, showed a large increase in the PL signal suggesting that the participation of defects in carrier relaxation is unlikely. Time resolved experiments could give direct information on the relaxation rates but they cannot unambiguously identify the mechanism. Photoluminescence Excitation (PLE) experiments map out the density of states (DOS) in higher dimensional structures where carrier relaxation is not inhibited but in self-assembled QDs, where the details of the relaxation processes are not well understood and there is a range of dot sizes and composition, the PLE

spectra are weak and not easily interpreted. Some studies have concluded that LO phonon features are[2,12,13] present in the PLE spectra while others have concluded that they are dominated by absorption into excited states of the dots[14,15]. However, in almost all of these studies the experimental data are obtained from samples with strongly overlapping states making analysis of the PLE spectra difficult. In addition, only as-grown structures with similar inter-level energies have been investigated and some method of modifying the electronic levels is desirable to resolve this issue[16-20].

EXPERIMENTAL DETAILS

The samples studied here were cut from the same wafer consisting of a single QD layer grown at a low growth rate (0.01 MLs^{-1}). Further details on the growth can be found in reference 21. Each piece was coated with 100nm SiO_2 and subjected to rapid thermal anneals at different temperatures between 600^0C and 900^0C[20]. Low temperature PL is obtained by exciting the samples with an argon-ion laser and dispersing the emitted light with a SPEX 1404 double grating monochromator and detecting with a cooled Ge diode using lock-in techniques. The PLE was excited with a Titanium sapphire laser tunable over the range 860 – 1000 nm.

RESULTS

Annealing induces interdiffusion of the In and Ga atoms resulting in larger dots with a lower average In composition and the ground state (GS) emission energy E_0 can be blueshifted by up to 350 meV. Under high excitation the energy spacing of the optical transitions, ΔE, can be measured and varies from 68 meV for the as-grown sample to 19 meV for the sample subjected to the highest temperature anneal. As reported elsewhere in these proceedings[22] there is a linear relationship between E_0 and ΔE that is attributed to conservation of indium atoms during annealing. The observation of strong level filling in the annealed samples demonstrates that the annealed samples retain their QD-like properties. Table I lists the values of E_0 and ΔE deduced from PL measurements.

Sample	Anneal (^{0}C)	E_0 (meV)	WL (eV)	FWHM (meV)	ΔE (meV)
A	-	1.047	1.429	24	68
B	675	1.191	1.432	30	43
C	700	1.248	1.432	28	36
D	750	1.342	1.446	15	26
E	780	1.393	1.463	10	19

TABLE 1: Annealing temperatures, GS (E_0) and WL energies, linewidths and separation of the optical transitions, ΔE, for samples A-E.

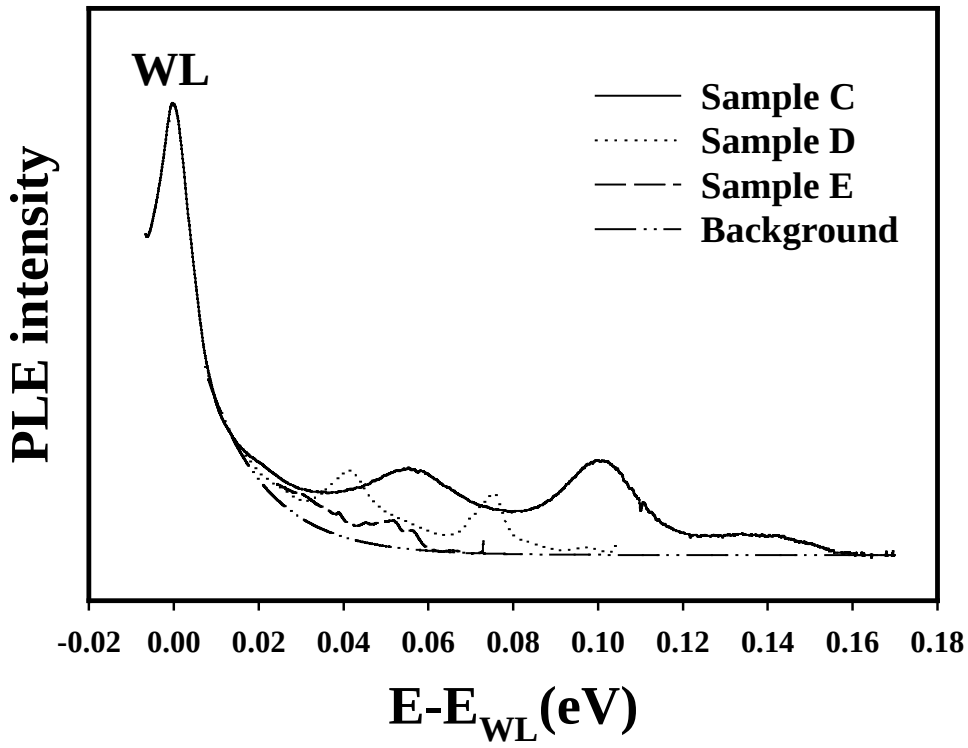

Figure 1:PLE signal from samples C, D and E. The intensities and energy have been normalized relative to the WL. The dashed-dotted line shows the background signal from the WL

Figure 1 shows the PLE (detected at the peak GS energy) signal for samples C, D and E, which have been shifted slightly so that the WL peaks are in coincidence and normalized in intensity. The general shapes of the spectra are remarkably similar and are dominated by a slowly reducing background signal shown by the dashed-dotted line that extends from the WL peak into the dot excited states. This feature is evident in the PLE spectra of all our samples and we believe it is common to all QD structures. If the WL were considered to be a more or less uniform quantum well a much narrower feature would be expected. However, the electronic structure of the WL will be affected by local variations of strain, thickness and composition leading to the broad absorption shown in figure 4. The presence of this absorption could have significant effects on the relaxation of carriers within the dots and such a process has already been proposed for single dots[23].

For each sample several relatively broad peaks are evident and these sit on the WL background. The separation of these features is similar to the values of ΔE listed in Table 1 suggesting that the PLE signals arise from absorption into excited states of the dots and they are not LO phonon resonances. Figure 2 shows these features in more detail for samples D and E (dotted and full lines respectively) with the background signal removed. The main features have

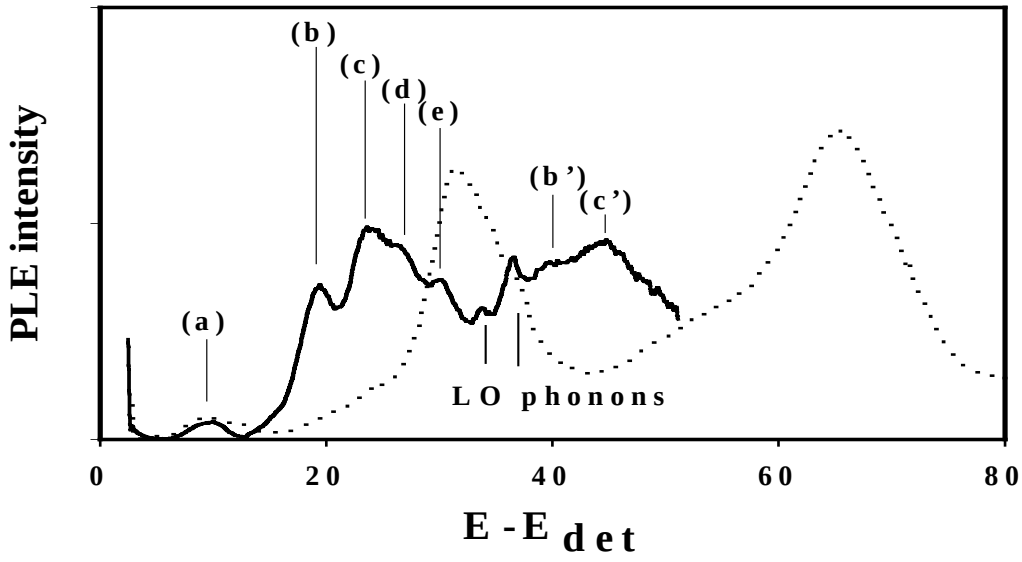

Figure 2: PLE signal for samples D (dotted) and E (full) with the background signal removed. The separation of the peaks is consistent with absorption by the excited states of the dots. The features (a)-(e) are attributed to absorption involving different hole levels.

already been assigned to absorption into excited states but the fine structure (labeled (a)-(e)) seen in the signal from sample E remains to be explained. There is also structure in the PLE spectrum obtained from sample D but this is not as clear.

Two sharp features lying roughly 34 and 36 meV above the detection energy, E_{det}, are assigned to Raman scattering. They are present as shoulders on the E_1 absorption peak of sample D. Returning to sample E, features (a)-(e) do not correspond to any known phonon in the InAs/GaAs system and we believe they arise from E_1-$H_{0\text{-}3}$ transitions. Figure 3 shows a schematic representation of the electronic levels that may exist in one of the dots in sample E. High excitation PL spectra show that there are four transitions corresponding to four-confined electron levels and at least four hole levels for samples D and E. The PLE signal in the region of the E_1-H_1 peak might then consist of contributions from the allowed E_1-H_1 transition (peak (c) in figure 2) and at least three "forbidden" transitions corresponding to the features (b), (d) and (e) shown in figure 2. If this interpretation is correct then similar features should be present in the PLE signal in the region of the E_2-H_2 transition. Thus the peaks labeled (b') and (c') in figure 2 are assigned to the E_2-H_0 and E_2-H_1 transitions. The relative strengths of the features will depend on the detailed nature of the wavefunctions while the separation will depend on the size and composition of the dots. The fine structure is not so clear in sample D and is not evident in the PLE spectra for sample C (figure 1) and it may be that the increasing inhomogeneous broadening in these samples is responsible. The separation of the features (b)-(c) etc. is roughly 2-3

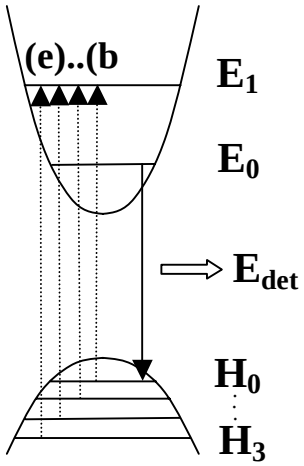

Figure 3: Schematic of the electronic levels in sample E. Transitions occur between E_1 and hole states H_0-H_3.

meV and this should be compared with ΔE=19 meV. This would imply an electron inter-level separation of 16 meV and a band offset ratio around 80:20.

The feature labeled (a) in figure 2 can be explained by a similar argument. Figure 4 shows the PLE signal from samples D and E close to the detection energy, E_{det}. Although there are no discernable features in the spectrum obtained from sample E, sample D exhibits three peaks labeled (c''), (d'') and (e'') and these are attributed to transitions involving E_0 and the H_1, H_2 and H_3 hole states respectively. Again, the relative intensities will be governed by the actual wavefunctions. Because of the strong scattered light signal it is not possible to detect the E_0-H_0 transition. From the energies of the peaks we can deduce that the separation of the hole states in sample D is 4-5 meV slightly larger, as expected, than those estimated for sample E.

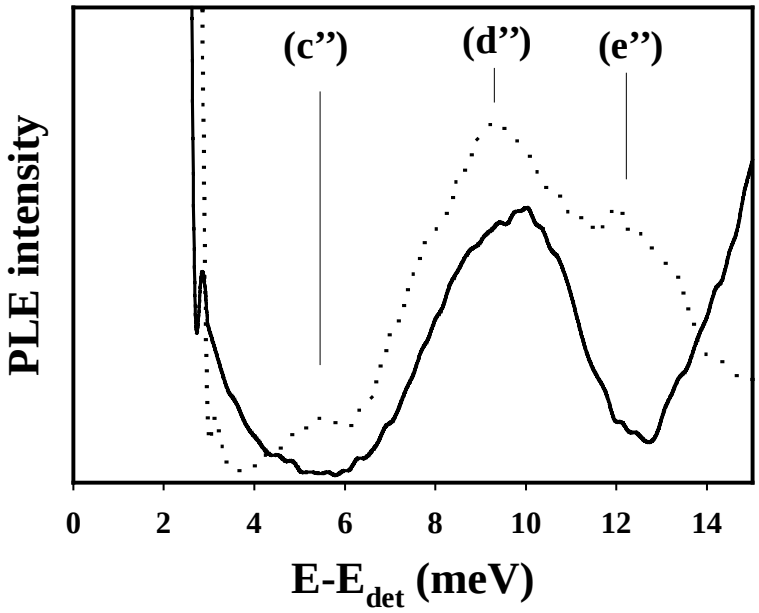

Figure 4: PLE signal from samples D and E close to E_{det} showing fine structure in the feature labeled (a) in figure 2. This structure is assigned to forbidden transitions involving the E_0 state and the H_1, H_2 and H_3 hole states.

CONCLUSIONS

We have shown that PLE spectra obtained from annealed low growth rate QDs reflect the density of states and are not the result of multi-LO phonon resonances. When the separation of the electronic states is less than the LO phonon energies fine structure is present in the PLE spectra which we attribute to forbidden transitions involving all the hole states.

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