Mat. Res. Soc. Symp. Proc. Vol. 642 © 2001 Materials Research Society

Radiative Decay and Carrier Relaxation in Annealed InAs/GaAs Self-Assembled Quantum Dots

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ABSTRACT

We have investigated the carrier relaxation and radiative decay in a series of annealed InAs/GaAs self-assembled quantum dots deposited at a low growth rate. The annealed samples retain their dot-like character, the ground state blueshifts and there is a decrease in the energy separation of the optical transitions from 68 meV to 19 meV. The photoluminescence (PL) transients have fast rise times consistent with rapid inter-level relaxation. Radiative lifetimes of the different states can be deduced from the transients providing they are normalised using time resolved photoluminescence. We believe that spin must be conserved during the inter-level scattering. As the dot composition and size changes the radiative lifetimes of the ground and first excited states decrease significantly and we attribute this trend to a combination of a shift in the emission energies and an increase in the overlap of the electron and hole wavefunctions.

INTRODUCTION

There have been a number of reports of the properties of self-assembled quantum dots (QDs) that are subjected to *in situ* or post growth annealing[1-4]. In all cases there is a shift of the emission to lower wavelength, a narrowing of the linewidth of the ensemble and a reduction in the separation of the electronic states. It should then be possible to tune the state separation of the annealed QD around the LO phonon energy. It is expected that carrier relaxation between the atomic-like levels will be inhibited if the inter-sublevel spacing is not equal to a multiple of the LO phonon energy (phonon bottleneck). In principle, time resolved studies should provide an insight into the carrier relaxation processes in QDs[5-9]. However, a problem in the analysis of decay transients is the treatment of "state blocking" where relaxation to the lower states is inhibited by the relatively long radiative lifetime of the ground state. In most cases the experimental data have been obtained from samples that exhibit large inhomogeneous linewidths. Under the conditions of high excitation intensity there are overlapping contributions from the ground and excited states making an accurate deconvolution of the signal difficult. In this paper we present PL, PL decay and time resolved PL (TRPL) measurements on a series of annealed samples grown at ultra low growth rate[10] which have narrow linewidths. It is generally accepted that self-assembled QDs correspond to the intermediate confinement regime where Coulombic effects are small compared with the confinement energies[11]. During the annealing process the dots increase in size and the inter-level energy separation decreases. Thus we might expect that annealed dots tend towards the weak confinement regime where there should be a strong dependence of the radiative lifetime with increasing dot size.

EXPERIMENTAL DETAILS

The wafer used in this investigation was grown using conventional solid source molecular beam epitaxy on an undoped (001) GaAs substrate and further details can be found in the paper by Le Ru *et al.* (these proceedings[12]). Cleaved pieces of the wafer were coated with 100nm of SiO_2 and subjected to isochronal (10s) anneals at temperatures between 600^0C and 850^0C. After annealing the SiO_2 layer was removed by dipping in HF for 30s. A control sample, which was capped but not annealed, did not show any shift in the emission wavelength after removal of the dielectric cap. PL measurements were made using an Ar^+ or Tsunami pulsed Ti-Sapphire laser (pulse width~1.5 ps) as excitation source. The luminescence was dispersed with a SPEX 0.85m double grating monochromator and detected with a Ge diode using lock-in techniques. A micro-channel plate (MCP) with an S1 response was used for the time resolved experiments. The system response is ~ 30 ps. Unfortunately the spectral range of the MCP does not allow measurements beyond ~1200 nm and consequently all data presented here correspond to a sample temperature of 12 K.

RESULTS

Figure 1 shows the low temperature PL obtained from all the samples under the conditions of high excitation power. There is clear excited state emission, which we take as evidence that the annealed samples retain their QD properties. The shift of the ground state (GS) emission is ~350 meV and the energy separation of the optical transitions, ΔE, varies from 68 meV in the as-grown sample (A) to 19 meV in the sample subjected to the highest temperature anneal (E).

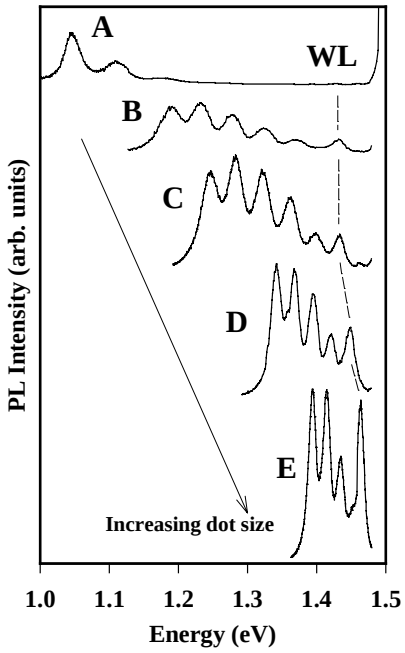

Figure 1: Low temperature PL obtained from the as-grown (A) and annealed (B-E) samples under high excitation.

Figure 2 shows the TRPL spectra obtained for sample C with ΔE~37meV. The signal is measured in a range ±50ps about the time shown on each spectrum. These "snapshots" allow the dynamics of the whole ensemble to be viewed. After 300ps the ground state is filled and this prevents relaxation from the upper states. Consequently there is a detectable signal from all the excited states. The ground state signal remains roughly constant for the next 1000ps and finally decays.

During this time the signal from the excited states decays sequentially from the highest to the lowest. This is entirely consistent with the picture of state blocking by the lower states. Note that the WL/CL[13] signal decays fastest consistent with fast carrier transfer to the dots and there are no resonances that might be

identified with LO phonons[7] in the spectra. This was true of all the samples investigated.

Figure 3(a) shows the decay transients for the sample C obtained at three excitation levels (350, 12 and 1 Wcm^{-2}). These data convey the same information as the TRPL spectra but allow a measurement of the radiative decays from the various states. Similar transients were obtained from all the samples. Comparing the rise times from all the annealed samples these are close to our time resolution (30ps) regardless of excitation level. This means that carrier relaxation is fast for all the samples investigated here and we find no evidence for LO phonon resonances. It may be that small variations in the relaxation times do exist but there is no evidence for a phonon bottleneck at least for the range of $47<\Delta E<19$ meV. We do see evidence of slowed relaxation (τ_{rise}~150ps) but *only* for the E_0 state of the unannealed sample with ΔE = 68 meV. Thus we can exclude contributions to τ_{rise} from capture from the barrier. This result would suggest that there is a dependence of the relaxation rate on ΔE but only when this becomes large. PL excitation studies presented elsewhere in these proceedings[14] (see Childs *et al.*) suggests that relaxation in annealed QD structures is aided by the presence of a *continuum*[15] extending from the WL. For the annealed samples the continuum extends to the lower QD states providing a rapid relaxation mechanism. An additional difference between the as-grown and annealed samples is the bi-exponential nature of the decays obtained from the excited states. This has been reported by several groups and will not be discussed in detail here. However, we believe that the bi-exponential behaviour is linked to the slower radiative decay of the ground state.

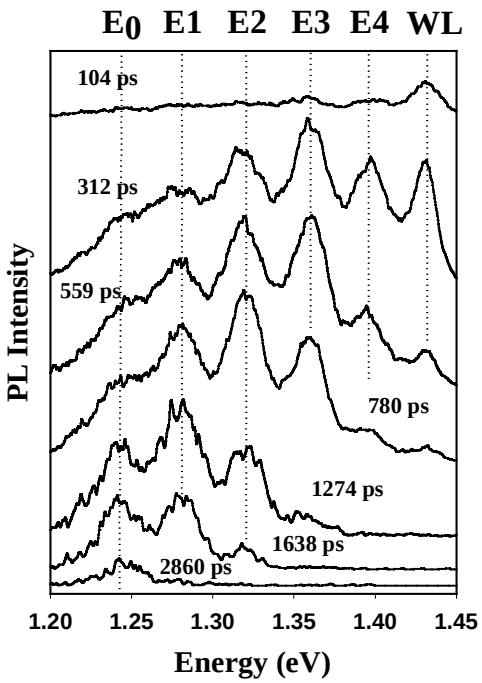

Figure 2 Time resolved PL obtained from sample C. The data were taken within a ±50ps window about the time indicated on each spectrum.

For the annealed samples the excited state transients can be fit to a single exponential decay. A good approximation to the radiative lifetime of the E_0 state can be obtained by measuring the slope of the transient at long times (>2ns) when the population of the excited states is depleted. Several groups have estimated this to be ~ 1ns for unannealed dots. Measuring the radiative lifetimes of the excited states from the transients is complicated by the relaxation from higher states and relaxation to lower states. At intermediate times when there is only substantial emission from the E_0 and E_1 we can assume for this three level system that:

$$\frac{1}{\tau_{meas}} = \frac{1}{\tau_1} + \frac{g_0}{\tau_0}$$

where τ_{meas} is the measured decay of the E_1 transition, τ_0 and τ_1 are the actual radiative lifetimes and g_0 is the degeneracy of the E_0 state. Inserting τ_{meas}=350ps and τ_0=620ps for sample C and assuming g_0=2 yields a negative value for τ_1. If instead we assume g_0 = 1 a much more realistic value for the radiative lifetime of the E_1 state of 800ps is obtained.

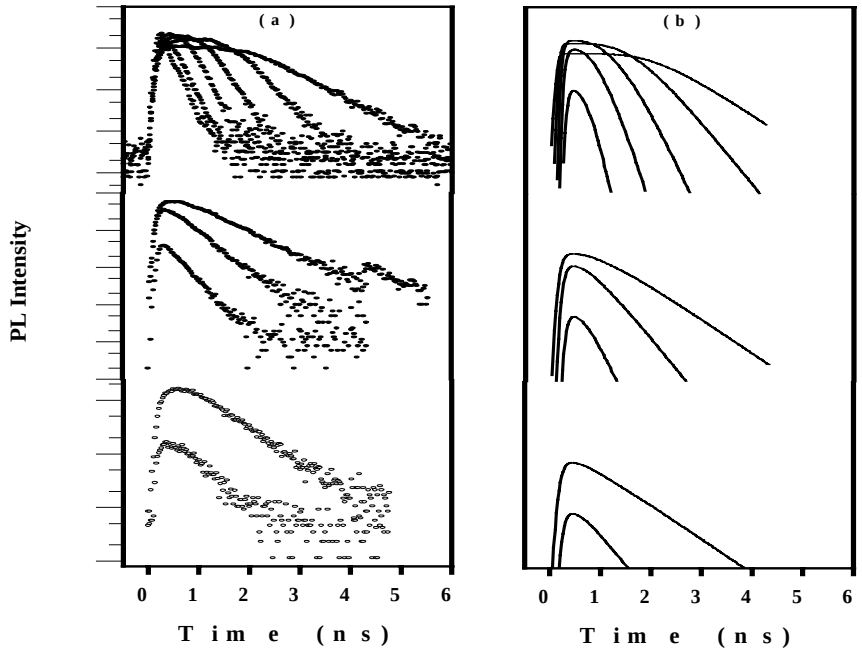

Figure 3 PL transients for sample C. Experimental data is shown in (a) for the E_0 and excited states at 350, 12 and 1 Wcm^{-2} (upper, middle and lower part of the diagram respectively). Note that at lower excitation levels the higher lying states are not detected. From these data, values of τ_0 and τ_1 can be deduced and used as input parameters to the random population model. The results of the model are shown on the right. For each excitation level the same radiative lifetimes are used and only the generation rate is changed.

Before making further comment on the degeneracy of the states we will present an alternative method of determining τ_1. At high excitation the dots are filled with carriers, which are replenished from the WL/CL as they recombine or are scattered to lower lying states. This leads to the plateau regions evident in the upper part of figure 3(a). During this time the emission from the E_0 and E_1 states is given by:

$$I_0 = \frac{g_0}{\tau_0} \text{ and } I_1 = \frac{g_1}{\tau_1}$$

Then the ratio of the intensities $R = I_1/I_0 = g_1\tau_0/g_0\tau_1$. An accurate measure of R requires normalisation of the PL transients both in terms of the detector response and the relative intensity of the signal from each state. This latter is achieved using the TRPL spectra shown in figure 2 and the data in figure 3 have been normalised using this method. This gives R=1.6 giving $\tau_1 = \tau_0/0.8 = 775$ps in good agreement with the method used earlier but which assumed $g_0 = 1$. Note that the assumption of $g_1/g_0 = 2$ does not contradict this argument. This analysis immediately raises the question of why the E_0 state behaves as if it had a degeneracy of 1. We believe that this is due to conservation of spin during relaxation. An exciton with a particular spin occupying the E_0 state will inhibit relaxation of excitons with the same total spin.

The transients from all the other samples can be analysed in a similar manner and we have checked for self-consistency by applying a random population model[16] (RP) using a

minimum of input parameters. It is clear from the TRPL and PL transients that inter-level relaxation is very fast compared with the radiative lifetimes of the states and we assume this to be infinitely fast for the model. We have already shown how we can obtain reasonable estimates for τ_0 and τ_1 and these are used as inputs. The radiative lifetimes of all the other states are adjusted to obtain a satisfactory fit. We can check for self-consistency by using the same radiative lifetimes and changing only the carrier generation rate G to generate fits for the data obtained at lower excitation rates. These fits are shown in figure 3(b) and are remarkably good considering the limited input parameters. Table 1 lists the radiative lifetimes of all the annealed samples used to obtain the fits.

Figure 4 shows τ_0 and τ_1 plotted against E_0. Both τ_0 and τ_1 decrease with annealing and consequently with increasing dot size and decreasing indium content. Plotting the values against the GS (E_0) emission energy shows a monotonic decrease in the radiative lifetimes. Figure 4 shows plots of τ_0 and τ_1 for all the samples. The lifetime is usually quoted as:

	A	B	C	D	E
E_0	800	650	600	550	490
E_1	920	783	720	611	563
E_2	889	903	812	689	653
E_3	1143	1083	941	917	N/A
E_4	1256	1300	1100	N/A	N/A

Table 1 Lifetimes for each transition used as inputs to the RP model.

$$\frac{1}{\tau} = \frac{e^2 E_P n \omega}{2 m_0 \varepsilon_0 h c^3} \left| \langle F_v | F_c \rangle \right|^2$$

where the symbols have their usual meanings. E_P is the Kane energy, which is taken to be 21.1 eV. The dotted line in figure 4 shows the effect of the GS energy (ω) on the lifetime. This is a result of the decreased indium concentration in the annealed dots that leads to a shallower confining potential.

It is clear that this accounts for much of the change in the lifetime for each state. However, the overlap of the wavefunctions may also have an influence on τ. Taking the difference between the dotted line and the experimental data to be due to changes in the overlap the inset in figure 4 shows a modest increase with annealing. The slight increase can be understood in terms of electron penetration of the barrier, which decreases with increasing dot size.

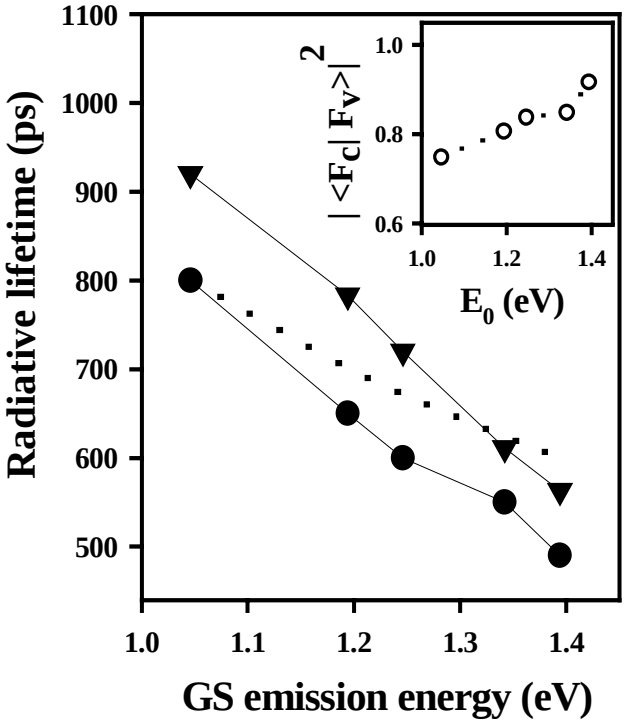

Figure 4 Values of τ_0 (circles) and τ_1 (triangles) used to fit the PL transients. The dotted line shows the influence of the change in GS energy while the inset

CONCLUSIONS

We have shown that the relaxation times in annealed QDs with different interlevel spacings are faster than 30ps. The radiative lifetimes decrease with increasing dot size and decreasing indium composition. Analysis of the PL transients normalised with TRPL spectra suggest that the ground state behaves as if the degeneracy is 1 rather than 2. We suggest that this is a result of conservation of spin during relaxation. There is a decrease in the lifetimes of the ground and first excited states with annealing which is mainly due to the change in emission energy of the GS.

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