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Superlinear Dependence of the Photoluminescence from InAs/GaAs Self-Assembled Quantum Dots

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ABSTRACT

The threshold current of lasers with InAs/GaAs quantum dot active regions is found to increase significantly above 250 K in contradiction to their expected “ideal” behavior. This would appear to be related to the loss of carriers due to thermal emission into the barrier or wetting layer. We have investigated the excitation density dependence of the photoluminescence at different temperatures in low growth rate quantum dots that emit close to 1.3 μm at room temperature. Similar experiments were performed on annealed samples where the barrier height is smaller and these demonstrate that the results are quite general. At low temperatures the photoluminescence signal increases linearly with the incident laser power. However, at higher temperatures where there is significant carrier escape a superlinear dependence is observed. Possible qualitative explanations for this phenomenon are given and discussed.

INTRODUCTION

Self-assembled quantum dots (QDs) are predicted to replace quantum wells as the active layer of many optical and electro-optical devices. Three dimensional confinement of carriers leads to unique properties such as a delta-like density of states and ultra-narrow, temperature independent, homogeneous broadening. In principle, QD lasers should have very low threshold currents and improved critical temperatures. The devices fabricated so far have not yet met these expectations and many issues remain to be understood. Among these, the behavior at high temperature is of great importance because most devices operate at room temperature or higher. Most studies [1-8] report a quenching of the photoluminescence (PL) intensity with increasing temperature. This is usually attributed to thermal escape of carriers from the dots into the barrier material [2,5-8], where they are lost through for example non-radiative recombination [4]. The dependence of the emission energy and the Full Width at Half Maximum (FWHM) have also been investigated and explained [2,6,7]. Fewer studies [5,8] report excitation density dependence studies at different temperature. This is particularly important for room temperature operation of lasers. The regime of very high excitation will mimic the behavior of the laser at high injection current. It is well known that for QDs under high excitation, state-blocking effects lead to saturation of the ground state (GS) emission and appearance of emission from the higher excited state. What happens under very low excitation is often overlooked, but should play a major role in the threshold current and critical temperature of the lasers. In this letter, we report measurements of the excitation density dependence of the PL intensity in the temperature range from 10 K to 300 K, in the regime of low excitation density. At 10 K, this dependence is linear as expected. For higher temperatures, where thermal escape dominates, this dependence becomes superlinear.

EXPERIMENTAL DETAILS

One sample (A) was grown using conventional solid source MBE but with the InAs layer deposited at a low-growth rate (≈ 0.01 ML/s). This results in emission close to 1.3 μm at 300 K (1.2 μm at 10 K), combined with a small FWHM [9]. The height of the barrier is taken to be the difference between the GaAs band gap and the emission energy of the GS, and is thus around 460 meV. This barrier is quite high compared to samples studied in previous works. To make sure our results were not dependent on the barrier height, we used Rapid Thermal Annealing (RTA) at different temperatures to produce 4 other samples. It has been shown previously that RTA induces Indium/Gallium interdiffusion, thus having the effect of blueshifting the GS energy, and therefore decreasing the barrier height [10,11]. Spectra of these samples are shown in Ref. 12. These annealed samples have barrier heights of 360 meV (B), 260 meV (C), and 170 meV (D), with GS emission at 1.151, 1.248, 1.342, eV at 10 K. PL spectra were obtained using an Ar^+ ion laser or an He-Ne laser for excitation above the barrier, and recorded with a 0.5 m grating monochromator and cooled Ge diode using standard lock-in techniques. In order to study the very low excitation density regime, the beam was not focused and had a diameter of around 1 mm at the sample surface. In this way, the intensity collected remains reasonable even though the density of excitation is much smaller than that conventionally employed using a focusing lens. Filters were used to decrease further the excitation. The experiments presented in the following have been carried out on all four samples and similar results were obtained for all of them. For the as-grown sample, the barrier is so high that the regime of strong thermal escape (and therefore strong quenching) is not really reached at 300 K. We will therefore present the results for sample C, where we can more easily probe the strong thermal escape regime. A more detailed description of the influence of the barrier height upon the temperature properties of the dots in these samples will be the subject of another publication.

RESULTS

Figure 1 shows the Arrhenius plots of the Integrated PL intensity **(**IPL**)** between 10K and 300K for the four samples. The shape of these curves is typical of the temperature dependence for QDs. Two main features can be observed on this plot. First, at high temperature, the curves tend towards a straight line, characteristic of an exponential quenching due to thermal escape from the dots. We can deduce an activation energy for each sample by measuring the slope. This activation energy matches the difference between the GaAs band gap and the ground state energy. Second, the quenching temperature increases with the barrier height and is close to room temperature for sample A. It is however interesting to note that although the quenching occurs earlier for small barrier heights, it is then slower. Extrapolating the plots, the intensity of sample D will be greater than that of sample C at around 340 K.

Figure 2 shows the Arrhenius plots of the Integrated PL intensity (IPL) between 100 K and 300 K for 4 different excitation densities. Conventional focusing lens was used to obtain the highest excitation density, estimated to be 10 W cm^{-2}, and which corresponds roughly to the excitation when a small signal from the first excited state becomes detectable. The three other sets of data were obtained without focusing, using filters to achieve excitation densities 20, 200 and 400 times smaller than in the first case. Only by not using a focusing lens is it possible to detect a reasonable signal at such small excitations. The curves shown have been normalized to have the same IPL at 10 K. The IPL remains constant between 10 K and 100 K in the three cases (not shown here). The shape of these curves is typical of the temperature dependence for QDs, showing an exponential quenching at high temperature. Measuring the slope at high temperature gives us the activation energy for thermal escape from the dots. These activation energies are equal in all four cases within the experimental errors. We derive a value of $E_a \approx 270$ meV, which matches the barrier height of this sample. It is not surprising to find the same activation energy in the four cases as it should not be dependent on the excitation density. However, as the excitation level decreases, the quenching of the IPL occurs at lower temperatures and the curves diverge as the temperature increases. If the dependence upon excitation density was linear, the three curves should be indistinguishable. This is the case between 10 K and 150 K, where the carriers are unable to escape from the dots and the dependence is linear (there are no state-blocking effects at such small excitation). For higher temperatures, where thermal escape becomes dominant, the divergence of these curves shows the dependence becomes *superlinear*.

To investigate this further, we made a more comprehensive study of the excitation density dependence of the IPL in the temperature range from 10 K to 220 K. The excitation density was varied from 500 to 16 mW cm^{-2}, giving a range of nearly 2 orders of magnitude. Note that all these excitation densities are in the regime where the number of electron-hole pairs per dot is much smaller than 1 and we do not see any emission from the excited states at 10 K.

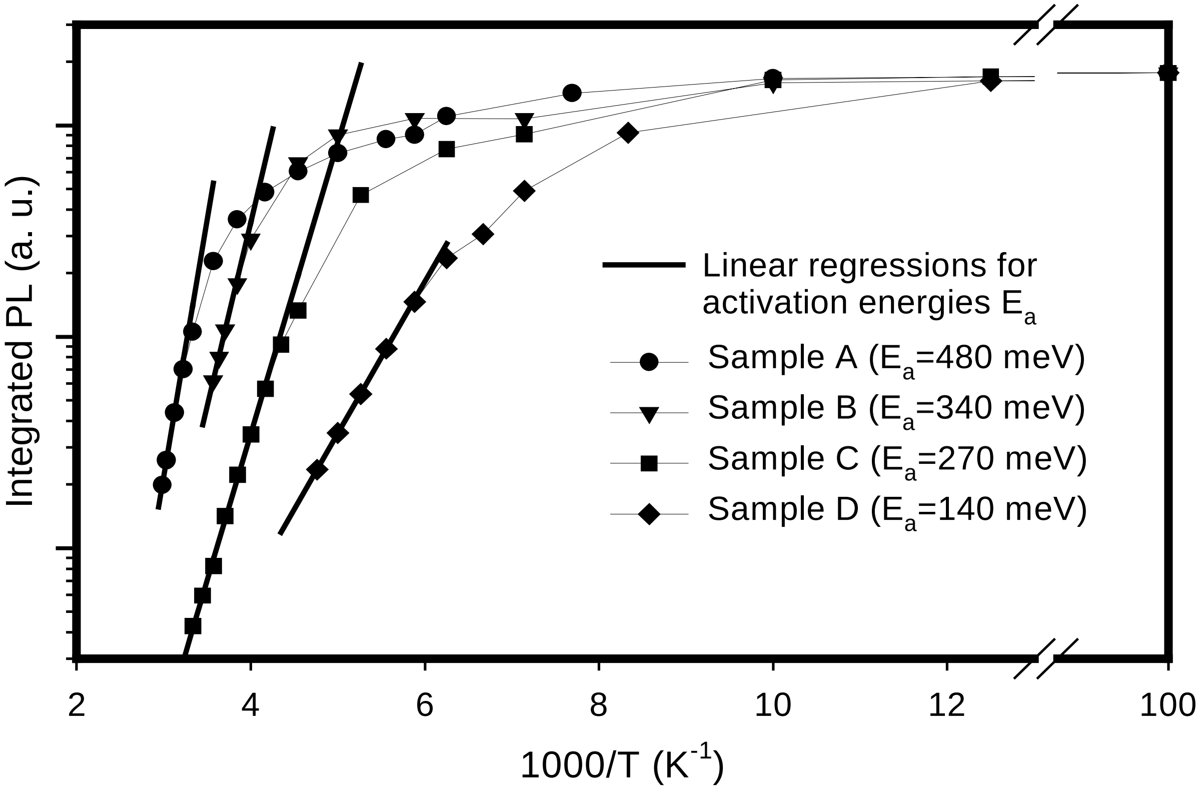

Figure 1: *Arrhenius plots of the temperature dependence of the integrated intensity from samples A (circles), B (triangles), C (squares) and D (diamonds). All curves are normalized to have the same intensity at 10 K. The straight lines are linear regression for the high temperature points. Their slopes give the activation energy of the thermal escape processes.*

To highlight the non-linearities, the excitation density is plotted against the IPL in a log-log plot at different temperatures. Figure 3 (a) shows the results obtained at 10 K, 130 K, 180 K, 200 K and 220 K. The straight lines are linear regressions. A purely linear behavior is characterized by a slope of exactly 1 and is what we observe for temperatures smaller than 150 K. However, at higher temperatures, the slopes increase, being close to 1.7 at 220K. The behavior is then superlinear, and can be seen more clearly on a linear plot shown in Figure 3 (b). Given the fact that the activation energy of the thermal escape is not dependent on the excitation level, the curves in Figure 2 should tend at high temperature towards parallel straight lines. In this regime, we should therefore in principle find a fixed law for the excitation dependence of the IPL at all higher temperatures. However, it is very difficult to give a definite conclusion on this issue, because in this regime the intensity is 3 to 4 orders of magnitude lower than those measured in conventional experiments, and the errors then begin to be significant.

Similar results were obtained for all the other samples. The only difference is the temperature when these effects start appearing. This temperature decreases with annealing temperature and corresponds for all of them to the characteristic temperature for which the IPL starts dropping (cf Figure 1). It shows that this effect is strongly related to the thermal escape of carriers in the barrier material.

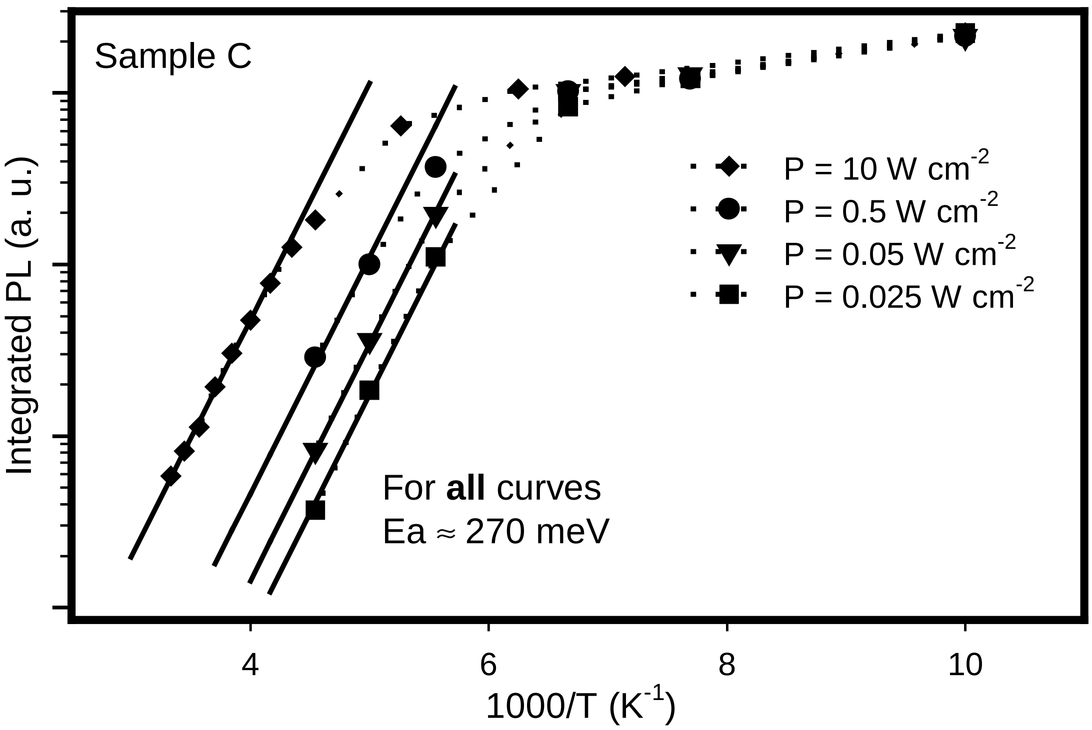

Figure 2: *Arrhenius plots of the temperature dependence of the integrated intensity from samples C between 100 and 300 K, at different excitation densities. The curves have been normalized to show the same intensity at 10 K.. The divergence of these curves reflects how the ratio between the intensities is changing with temperature. The straight lines are linear regression for the high temperature points showing that the activation energies are equal.*

DISCUSSION

At this stage, we can only speculate on the reasons for this superlinear dependence. We believe it is a characteristic of all QD samples under conditions of low excitation and high enough temperature. All the models available in the literature assume a linear dependence with excitation and cannot explain this behavior. We showed that the superlinearity was closely related to the regime of strong thermal escape, so we need to look for an explanation either in the way carriers are captured and emitted in the barrier, or in the way they are lost once in the barrier. We propose three possible mechanisms:

- Saturation of losses in the barrier: If part of the losses can be saturated when the population in the barrier increases, it could lead to a superlinear dependence at intermediate excitation when the losses start to be saturated. However, we would expect to have a linear dependence at smaller and larger excitations.
- Uncorrelated recapture of electrons and holes: If electrons and holes which escape from the dots are recaptured independently, then the probability of recapturing one electron and one hole in the same dot would vary quadratically at very low excitation leading to a superlinear dependence.
- Auger processes: Although Auger-assisted capture is thought to be negligible at low temperature and low excitation, it could become dominant at higher temperature when the carriers escape from the dot and will end up in the barrier material. Using values of the

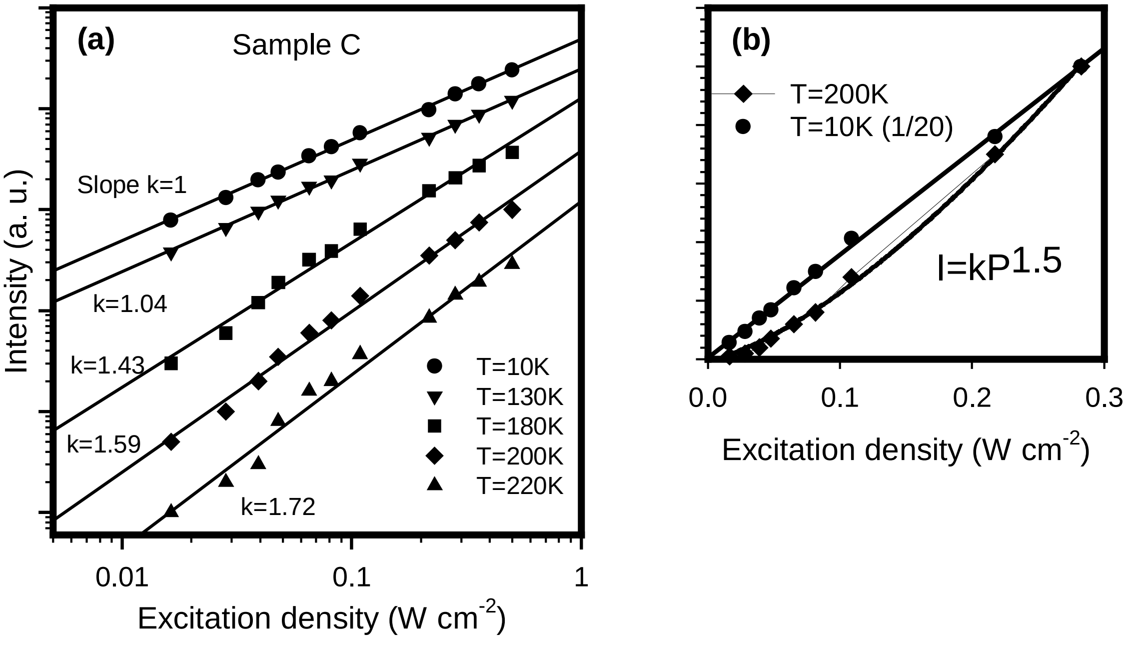

Figure 3: *(a) Log-Log plot of the dependence of the IPL with excitation at different temperatures for sample C. The solid lines are linear regressions, the slopes of which (k) characterize the degree of superlinearity. k=1 for a linear dependence, and slopes larger than 1 show a superlinear dependence. (b) Same plots on a linear scale for T=10K and T=200K.*

Auger coefficients measured experimentally [13], we see that Auger-assisted capture can be dominant for a population as low as 10^8 cm^{-2} in the wetting layer. Populations of this order are easily reached even at very low excitation as soon as the carriers escape thermally from the dots. The dominance of Auger-assisted capture at high temperatures could account for the superlinearity observed.

We cannot at this stage decide which of these effects is dominant. A more detailed discussion of this including modeling of the results will be the subject of another publication.

CONCLUSION

We have studied the excitation density dependence of the PL intensity at different temperatures in a set of QD samples with different barrier height. The dependence was linear at low temperature as expected. However, this dependence becomes superlinear as we increase the temperature. Comparing the data from different samples, we showed that the superlinearity appeared only in the regime of thermal escape from the QDs (when quenching of the emission is observed). Several possible explanations have been proposed based on our observations. This regime of low excitation at room temperature should be particularly relevant in the study and determination of threshold currents in QD laser structures.

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