

Luminescence Enhancement by Hydrogen Passivation in InAs/GaAs Self-Assembled Quantum Dots

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ABSTRACT

There is a consensus that point defects may be present in quantum dots and these may influence the efficiency of light emitting devices with quantum dot active regions. In addition the design of these structures requires growth of GaAs cladding layers at substrate temperatures that are less than optimum thus increasing the possibility of incorporating non-radiative centers close to the dots. It is well known that atomic Hydrogen passivates shallow and deep defects in bulk and quantum well material, and therefore may be beneficial in quantum dot structures. Hydrogen was introduced into the samples via an inductively coupled plasma and the emission compared with a control sample across the temperature range 10-310 K. The passivation treatment appears to be benign and there is no significant change in the emission energy or the linewidth. Relatively small enhancements in the emission intensity ($\times 2$) are measured at low temperatures but this increases by an order of magnitude at ambient temperatures. The enhancement is attributed to passivation of non-radiative defects in the surrounding GaAs barrier and wetting layer. In principle, by improving the spontaneous radiative efficiency in QD structures, hydrogen passivation offers the prospect for ultra-low laser thresholds.

INTRODUCTION

The use of Quantum Dots (QDs) is predicted to improve substantially the performance of optoelectronic devices such as lasers. However, devices fabricated so far have not achieved all the expectations. The presence and role, if any, of defects in QD structures have not been assessed in detail, but they are likely to have an impact on the performance of devices [1,2]. Hydrogen can be introduced from a plasma [3], ion source [4], or during Molecular Beam Epitaxy (MBE) [5] and is known to passivate shallow and deep defects in bulk and quantum well materials [6] and has been shown to improve the radiative efficiency [7]. The passivation of defects in QD structures is desirable not only for the improvement of device performance but also from a fundamental point of view to understand the role of defects in the optical properties. A recent study demonstrated the possible destructive effects of high energy protons on the emission from QDs [8]. However, we have shown that Hydrogen passivation could also lead to dramatic enhancement in the radiative efficiency of QDs [9]. Here, we extend that study to show that Hydrogen leads to a substantial passivation of defects in both the Wetting Layer (WL) and the GaAs matrix adjacent to the dots.

EXPERIMENTAL DETAILS

The samples were grown using conventional solid source MBE on a (001) GaAs substrate. After deposition of a 100 nm GaAs buffer layer and a six-period AlAs/GaAs superlattice at a substrate temperature (T_s) of 580°C, T_s is reduced to 500°C. At this low temperature, 10 nm of GaAs, the QD layer and another 10 nm of GaAs are grown. Then T_s is ramped up to 580°C for growth of another superlattice and deposition of a final 50 nm of GaAs. In the first sample (A), the QD layer consists of 2 ML of InAs deposited at a low growth rate (≈ 0.01 ML/s) resulting in relatively large QDs (around 45 nm across and 8 nm high before capping), emitting around 1.3 μm at room temperature with a narrow linewidth of 25 meV [10]. The two superlattices were grown to isolate the QD layer from the surface, therefore avoiding any influence of the surface on our results. To check that the superlattice had no effect on the results either, another sample (B) was grown without the superlattice. The growth rate of the QD layer was also slightly higher to obtain emission at a shorter wavelength (1.24 μm at room temperature). Pieces of each wafer were then subjected to a Hydrogen plasma treatment at 280°C for one hour. Part of the sample was masked by a piece of undoped GaAs substrate to provide a control region. The emission from the control (masked) region was indistinguishable from the as-grown sample. Photoluminescence (PL) was measured using a He-Ne laser for excitation. The detection system consisted of a SPEX 1404 double grating monochromator and cooled Ge diode using standard lock-in techniques.

RESULTS

Figure 1 shows the spectra obtained at 10 and 300 K from the passivated and control parts of sample A. We see that the shape and position of the spectra are unchanged after passivation,

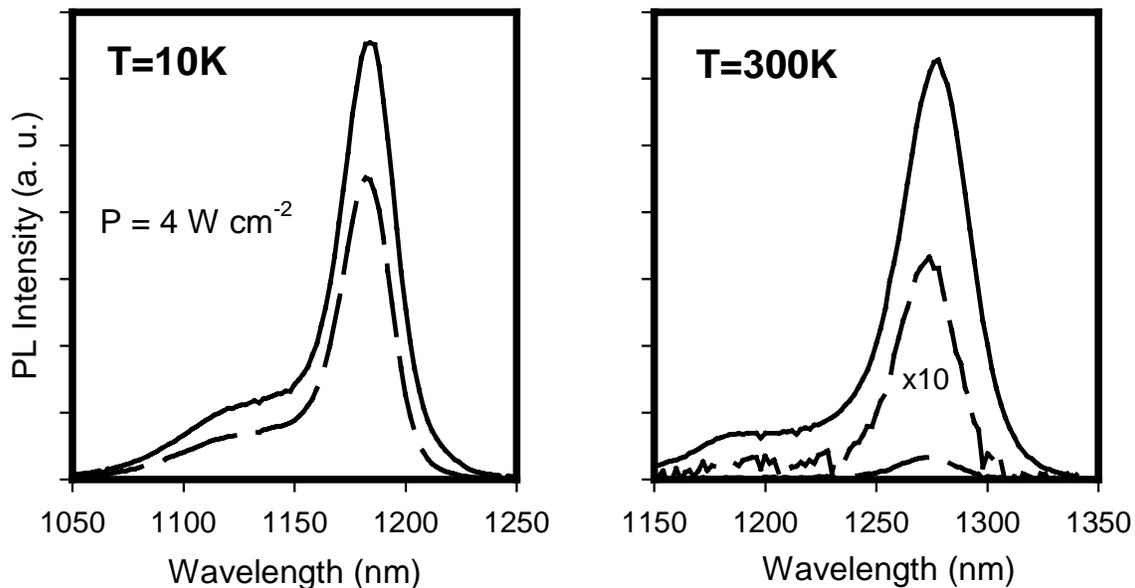


Figure 1. Spectra of the passivated (solid line) and control (dashed line) samples at low and room temperature for sample A. The shape and position of the spectrum are unchanged after passivation. The intensity is increased by a factor of 1.6 at 10 K, and ≈ 20 at 300 K.

showing that the Hydrogen had no effect on the electronic structure of the dots. However, the intensity of the PL from the passivated region is enhanced by a factor of 1.6 at 10 K. This enhancement is much stronger at room temperature, being close to 20. Similar results were obtained from sample B.

To understand why this enhancement is so temperature dependent, we carried out a more systematic study of the temperature dependence of the integrated PL intensity. The results are shown as Arrhenius plots in Figure 2. The shape of the Arrhenius plot obtained from the control part is typical of what we expect from QDs. However, the curve obtained from the passivated sample is different in several respects. First, we observe a slight decrease in the intensity as the temperature increases from 10 to 100 K. This results in the emission from the passivated and masked regions being identical for intermediate temperatures (between 100 and 150 K). However, as we increase the temperature further, the intensity from the passivated region remains roughly constant whereas that from the control sample is quenched. This effect results in a large increase in the enhancement ratio. Eventually, at higher temperatures, the emission from the passivated sample is also quenched. The quenching is exponential and is known to be due to thermal escape of carriers from the QDs into the barrier material. Not surprisingly, because the electronic structure was not affected by the treatment, we see that the activation

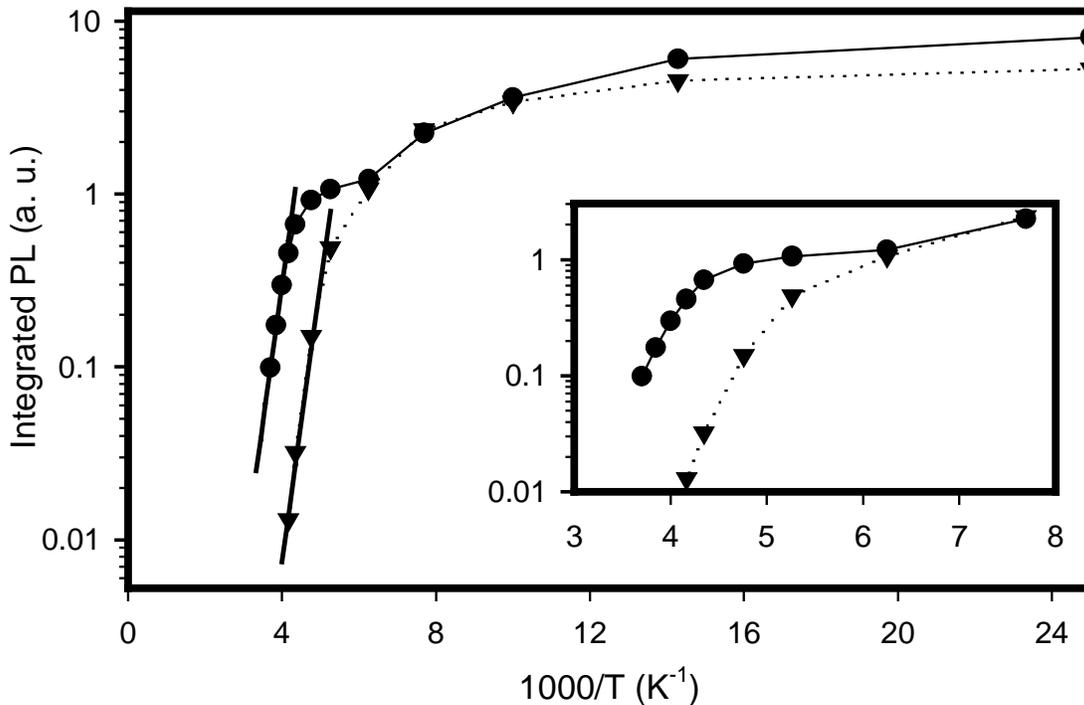


Figure 2. Arrhenius plots of the dependence of the integrated PL intensity for the passivated (circles) and control (triangles) parts of sample B. The excitation density is 4 W cm^{-2} . The solid lines at high temperatures are a linear regression obtained from the last few points. They show that the activation energy is the same in both parts. The inset shows a zoom at high temperatures showing the anomalous behavior of the passivated sample at intermediate temperatures.

energy is the same for both passivated and control parts, with a value equal to the energy difference of the barrier and QD ground state emission. What is surprising is the anomalous behavior of the passivated sample at intermediate temperatures, which results in the quenching of the PL at a much higher temperature than for the control sample. This effect is highlighted in the inset of Figure 2. Similar results are obtained for sample A, but the confinement potential is deeper in this sample (lower ground state energy), and so the quenching occurs at higher temperature even in the control sample. For the passivated sample A, the quenching has hardly started when we reach room temperature.

Elsewhere in these proceedings [11], we show that the dependence of the emission intensity from QDs with excitation density is superlinear at high temperatures where the quenching occurs. This is true before passivation and remains true after passivation. However, because the quenching occurs later in this case, the superlinearity also appears at higher temperature. This effect implies that the enhancement ratio is also excitation dependent at a given temperature. Figure 3 summarizes the measured enhancement ratios at different temperatures and excitation densities. We see that the strongest benefits of the passivation are observed at high temperatures and low excitation densities. Enhancement by two orders of magnitude can be measured before the emission of the control sample drops below the detection limit.

Finally, Figure 4 compares the wetting layer emission at 10K from the passivated and control samples. The intensity of the signal in the passivated sample is enhanced by a large factor of at least 20. Moreover, we observe a broadening of the peak towards the higher energies.

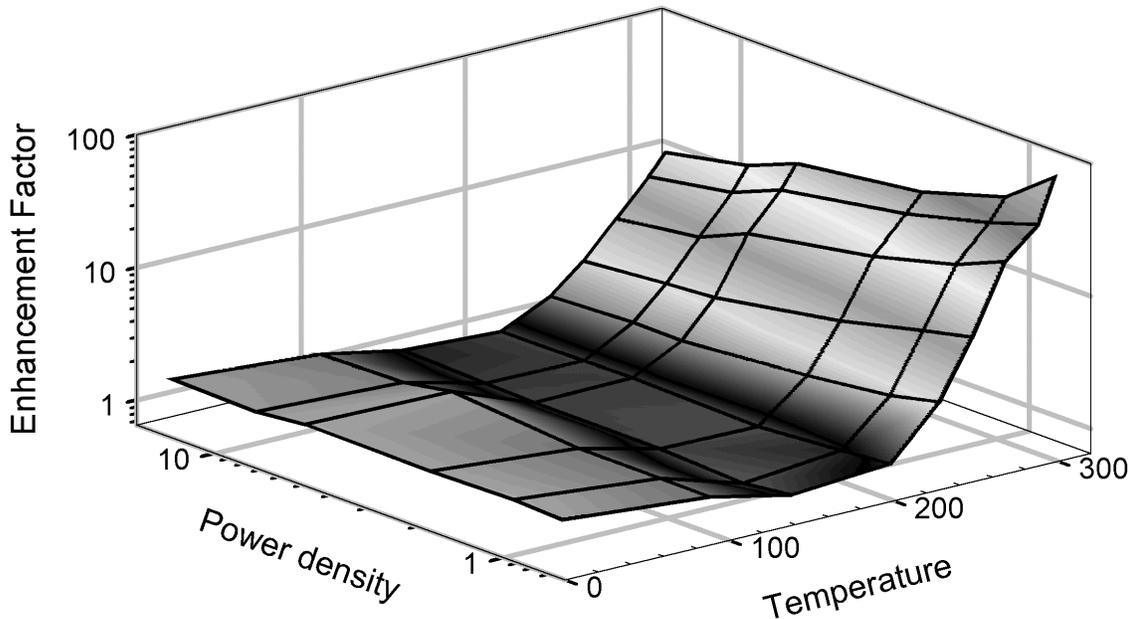


Figure 3. Plot of the enhancement factor R (ratio of the intensities of the passivated and the control sample) at different temperatures and excitation densities. R is close to 1.6 at 10 K and decreases to 1 at intermediate temperatures for all excitations. However at higher temperatures, R is strongly dependent on the excitation density. Enhancement factors of the order of 100 can be observed at high temperature and low excitation.

This indicates that the population of carriers in the WL is much larger. The signal from the GaAs band edge (at 1.51 eV) is also enhanced by two orders of magnitude (not shown here). These large enhancements in the WL and GaAs PL signals are attributed to a reduction in the rate of non-radiative losses. This is consistent with the passivation (by Hydrogen) of, as yet, unidentified non-radiative defects.

DISCUSSION

Analysing the experimental results, we can attempt to determine the effect of Hydrogen passivation on a QD sample. Because we observed the same effects on both samples A and B, we can exclude any influence of the surface or the interfaces of the superlattice. In the previous paragraph, we underlined that the most likely effect of Hydrogen was to passivate defects in the barrier material (WL and GaAs). Growth of the QD layer and subsequent capping requires a relatively low substrate temperature (500°C), which is not the optimum growth temperature for GaAs. In many cases, including ours, a thin buffer layer is also grown before the dots at this

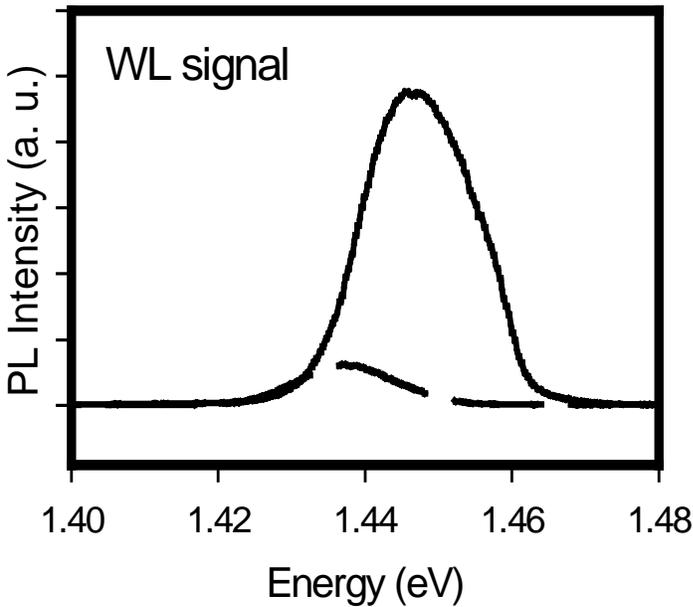


Figure 4. Photoluminescence of the Wetting Layer from the passivated (solid line) and control (dashed line) parts of sample B at 10K. After passivation, the intensity is strongly increased and the emission is broadened on the high energy side, indicating a higher population of carriers.

temperature. Therefore, the QD layer is embedded in a GaAs matrix which is likely to contain defects. It is possible that the passivation neutralizes efficiently some of those defects. We do not believe that Hydrogen passivates defects directly in the QDs. If this was the case, then we would expect a similar enhancement of the QD emission at all temperatures. In addition, assuming passivation of defects in the barrier material provides a plausible explanation for the temperature dependence of the enhancement. Carriers are captured very rapidly into QDs at low temperature. The presence of losses in the surrounding material has therefore little effect and the benefits of passivation are small. However, as the temperature increases, the balance between thermal escape from the dots and possible recapture leads to an increasingly longer effective capture time. Carriers then remain much longer in the barrier and any loss mechanisms can then have a strong effect on the overall QD intensity. Losses in the barrier therefore play a major role only at higher temperature. To a first approximation, the quenching will start at a temperature when the effective capture time becomes comparable to the

temperature. Therefore, the QD layer is embedded in a GaAs matrix which is likely to contain defects. It is possible that the passivation neutralizes efficiently some of those defects. We do not believe that Hydrogen passivates defects directly in the QDs. If this was the case, then we would expect a similar enhancement of the QD emission at all temperatures. In addition, assuming passivation of defects in the barrier material provides a plausible explanation for the temperature dependence of the enhancement. Carriers are captured very rapidly into QDs at low temperature. The presence of losses in the surrounding material has therefore little effect and the benefits of passivation are small. However, as the temperature increases, the balance between thermal escape from the dots and possible recapture leads to an

lifetime in the WL or barrier material. Because the concentration of defects is smaller in the passivated samples, the lifetime in the barrier is longer, and this explains why the intensity persists to a higher temperature. At even higher temperatures, the carrier lifetime in the barrier becomes much shorter than the effective capture time in both the control and passivated samples, and we observe the exponential quenching characteristic of thermal escape over the barrier (regime of strong quenching). The fact that the effects of passivation are strong only at high temperature demonstrates the great importance of the quality of the barrier material in the room temperature emission of QDs and consequently in many device applications.

From this discussion, we can note that we observed no evidence of passivation of defects in the dots themselves. The most probable explanation for this is that no point defects are actually present in the dots. It is however interesting to note that, even when the dots are free from defects, their properties at room temperature can still be highly influenced by the presence of defects in the surrounding material.

CONCLUSION

We have studied the effects of Hydrogen passivation on the luminescence efficiency of quantum dot samples. Although small enhancements are observed at low temperature, much larger improvements are obtained at room temperature (up to x100). We attribute these results to passivation of defects in the barrier material. Our conclusions highlight the importance of the defects for the room temperature emission of quantum dots.

ACKNOWLEDGEMENTS

One of us (ECLR) acknowledges financial support from the Ministère de l'éducation Nationale, de la Recherche et de la Technologie, France.

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