Luminescence enhancement from hydrogen-passivated self-assembled quantum dots

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We have measured a large increase (by a factor of up to 50) in the room-temperature emission of InAs/GaAs self-assembled quantum dots subjected to a hydrogen-passivation treatment. Smaller enhancements were measured at low temperatures. We tentatively attribute the improved optical signal to passivation of defects within the GaAs matrix and wetting layer adjacent to the dots. Annealing studies show that these benefits are lost following annealing at temperatures above 600 °C for 5 min. © *2000 American Institute of Physics.* [S0003-6951(00)03242-3]

There is continuing interest in the fundamental properties of InAs/GaAs self-assembled quantum dots (QDs). The interest is driven, in part, by the prospect of low threshold lasers and light-emitting diodes (LEDs) operating at the important telecommunications wavelengths. An aspect of their behavior, which has not been extensively investigated, is the role, if any, of inadvertent impurities and point defects on the emission properties. The presence of such defects is plausible given the complicated growth mode and the rather low temperatures used to deposit the GaAs barriers adjacent to the dot layer. Sercel[1] has proposed that the presence of deep traps such as M1 could provide an efficient relaxation path for electrons through multiphonon-assisted tunneling through the trap. Metastable behavior of the energy levels in QDs induced by spatially localized defects in the vicinity of the dots, similar to the *DX* center has been reported.[2] Other groups[3,4] have suggested that deep level defects may be responsible for the relatively low critical temperatures of laser structures. The presence of deep levels is often cited as a reason for poor optical quality in epitaxial GaAs/AlGaAs structures[5] but some studies have shown that hydrogen can have beneficial effects yielding higher radiative efficiencies.[6] Hydrogen can be introduced from a plasma,[7] ion source,[8] or during molecular beam epitaxy (MBE) growth[9] and is known to neutralize or passivate shallow and deep defects and impurities in GaAs and other III–V semiconductors.[10] There has been a recent report of the destructive effects of high energy protons on QDs[11] but none, to our knowledge, concentrating on the possible positive effects of hydrogen passivation. In this letter we report the effects of hydrogen passivation on the luminescence of InAs/GaAs self-assembled QDs. We find a significant improvement in the emission intensity, which we attribute to passivation of defects in the adjacent GaAs matrix and/or wetting layer (WL). The enhancements are temperature dependent being much larger at room temperature where the effects may be beneficial to laser performance. They are also excitation density dependent, being more pronounced at low excitation.

The sample studied here was grown on a semi-insulating (001) GaAs substrate by solid source molecular beam epitaxy. The QD layer consists of 2 ML of InAs deposited at a low growth rate of 0.01 ML/s resulting in relatively large islands that exhibit room temperature emission around 1.3 μm with a narrow linewidth of 25 meV.[12] A six-period AlAs/GaAs superlattice was also included 20 nm away on either side of the dot layer, thus avoiding any influence of the surface states. Pieces of the wafer were then subjected to a hydrogen plasma treatment at 280 °C for 1 h. Part of the sample was masked by a piece of undoped GaAs substrate to provide a control region. A second control sample was annealed at 280 °C for 1 h but without the plasma. The emission from both control samples was the same and was indistinguishable from the as-grown sample. Photoluminescence (PL) was measured for all the samples using a He–Ne laser for excitation. The detection system consisted of a SPEX 1404 double grating monochromator and cooled Ge diode.

Figure 1 compares the photoluminescence signal at 10 and 300 K for the passivated and control samples for an excitation density of 4 W/cm^2. Although a small redshift of around 3 meV is detectable in the emission energy of the passivated sample, the spectral shape is unchanged. We conclude that the treatment did not have any significant effects on the electronic structure of the dots. However, there is an enhancement by a factor of ≈1.6 in the emission intensity at

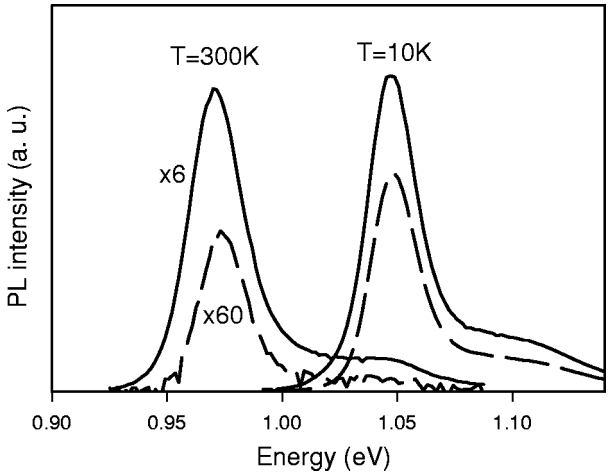

FIG. 1. PL spectra of the control (dashed line) and passivated (solid line) samples at 10 and 300 K. The enhancement factor is 1.6 at 10 K and increases to ≈20 at 300 K.

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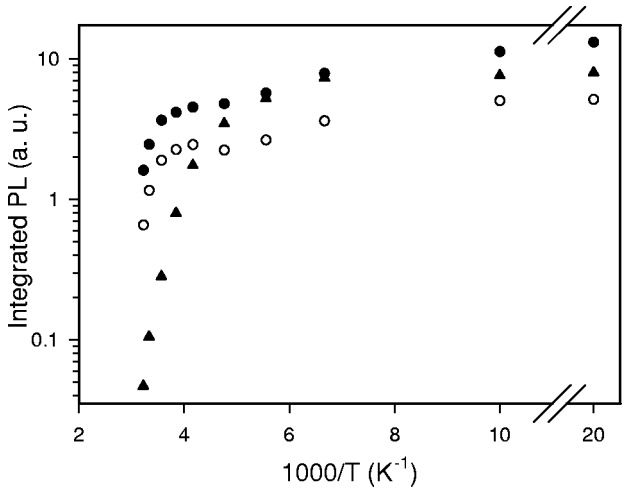

FIG. 2. Arrhenius plots of the temperature dependence of the integrated PL emission from the control (triangles) and passivated (circles) sample between 50 and 310 K at an excitation density of 4 W/cm^2. Empty circles show the data for the passivated sample at 1.5 W/cm^2.

low temperature, which increases to ≈20 at room temperature. This factor is even larger at higher temperature. The enhancement at room temperature is also dependent on the excitation level with factors up to 50 measured for lower excitation. At low temperature the enhancement is independent of the excitation level over several orders of magnitude. An increase in the low temperature emission in the region of the GaAs band gap [free exciton and $(e,C°)$] is also observed for the passivated sample. However, these signals become undetectable above 50 K and it is not possible to determine whether the enhancement persists to higher temperatures. Nevertheless, an increase in the band-edge signal implies a decrease in the concentration of defects in the GaAs, which we assume to exist in the layers grown at the lower temperature. This would increase the number of carriers transferred to the dot layer at low temperature, and can thus account for the enhancement observed at 10 K.

To investigate the mechanisms of the enhancement, we studied the integrated PL intensity (IPLI) in the temperature range 10–310 K for both samples. Figure 2 shows the Arrhenius plot of the IPLI signals. The slopes of the lines at high temperatures yield activation energies of 400 and 460 meV for the passivated and control samples, respectively. The activation energies are similar and are close to the energy difference between the QDs ground state (GS) emission and the GaAs band gap. The reduction in the luminescence is usually attributed to thermal escape into the barrier material followed by loss (for example by nonradiative recombination) in the barrier.[13] Because the electronic structure was not changed by the passivation treatment, it is not surprising that the activation energy for thermal escape is the same in both samples. However, we observe that the temperature at which quenching occurs is higher for the passivated sample. This results in larger enhancement factors at all higher temperatures. We also note that the intensity of the passivated sample drops slowly from 10 to 130 K and is then nearly equal to that of the control sample which remained stable until 130 K. Then the passivated sample exhibits an anomalous behavior between 170 and 250 K. The Arrhenius plot is relatively flat and a significant drop in the intensity occurs only above 280 K. Even when the excitation density for the passivated sample is reduced so that the IPLI is smaller than that of the control sample at 10 K (see Fig. 2), the emission is stronger at room temperature.

At this stage we do not have a comprehensive understanding of these results but we can give a qualitative explanation. We believe the passivation treatment decreases the concentration of defects, for example, nonradiative centers, in the GaAs barrier and also perhaps in the WL. Carriers are known to be captured very rapidly into QDs[14] at low temperature, and the presence of losses in the surrounding material therefore has little effect. The benefits of the passivation treatment are thus small. However, as the temperature increases, the process of thermal escape from the dots and possible recapture leads to a much longer effective capture time, which increases further with temperature. The 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 lifetime in the WL or barrier material. This lifetime should include all the possible loss channels: radiative recombination, nonradiative recombination, trapping mechanisms, surface recombination for near surface dots, etc. 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. However, for even higher temperature, the 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 a thermal escape over the barrier (regime of strong quenching).

To understand the fact that the enhancement factor was strongly dependent on the excitation density at room temperature, we carried out a more systematic study of the excitation level dependence of the PL at different temperatures. This dependence is linear at 10 K, but becomes superlinear at higher temperature when the PL is quenched. The superlinearity is particularly strong at low excitation and high temperature in the regime of strong quenching. This behavior was observed in all the QD samples we studied (before passivation), and also in strained InGaAs quantum wells. We therefore believe it is a general property of QD samples. Because this superlinearity appears at the same temperature as the PL is quenched it must be related to what happens to the carriers when they escape. Possible explanations are the saturation of some losses with increasing excitation, and the independent capture of electrons and holes at high temperature. The superlinearity is also observed for the passivated samples, but because the quenching occurs later, the superlinearity appears at higher temperature. Therefore, at room temperature the intensities of the two samples have a different dependence on the excitation density, the control sample being more superlinear. This results in an increase of the enhancement factor with decreasing excitation levels.

Enhancement of the luminescence of QDs is highly desirable and if maintained under the conditions of current injection would lead to more efficient light emitters. The fact

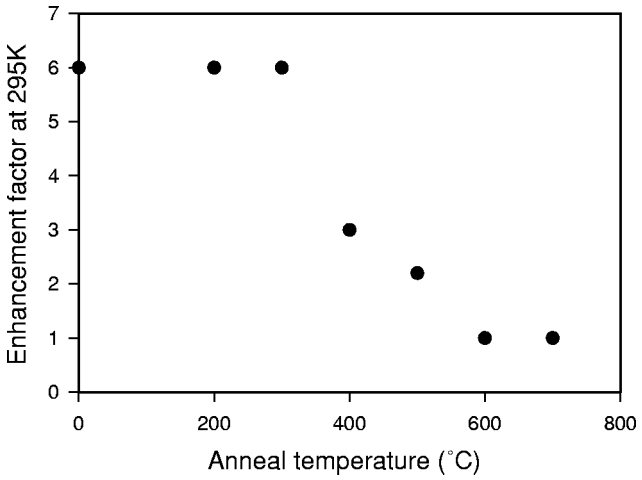

FIG. 3. Enhancement factor in the integrated emission of the passivated sample at 295 K compared to the control sample following an annealing treatment of 5 min at different temperatures. The excitation density is fixed at 16 W/cm^2.

that the enhancement is much larger at lower excitation density and room temperature is promising for low threshold QD lasers with high critical temperatures. However, previous studies of the effects of hydrogen on bulk or quantum wells have shown that the benefits can be lost during subsequent processing at high temperatures. Consequently, we have subjected a passivated sample to isochronal (5 min) anneals at different temperatures. To characterize the benefits of the treatment, we measured the enhancement factor of the PL after each anneal under the same excitation density at room temperature. Figure 3 shows a plot of this factor as a function of anneal temperature. As expected, the ratio decreases above 300 °C and the peak intensities of the passivated and control samples are equal after a 5 min anneal at 600 °C. Similar annealing behavior has been reported for passivated DX centers in AlGaAs.[7] This behavior is likely to limit the beneficial effects of hydrogen passivation if the temperatures of device processing are high and further work is in progress to determine the effects of hydrogen passivation in device structures. The escape of hydrogen from the sample is a thermally activated process, and this study shows that the effect of hydrogen should persist for long times if the devices are operated close to room temperature. Following the 600 °C anneal the sample loses two characteristic properties of the passivated sample presented previously, namely the slight shift in the emission energy, and the strong GaAs free exciton signal at 10 K. This confirms that these properties were a direct result of the presence of hydrogen in the sample.

In conclusion we have demonstrated large enhancements in the emission intensity of InAs/GaAs self-assembled quantum dot structures by the injection of atomic hydrogen. This treatment passivates defects in the GaAs barrier and possibly in the WL. The hydrogen can be removed by extended anneals at temperatures above 600 °C, but is stable at room temperature. This treatment is a good candidate to improve the threshold and critical temperature of quantum dot lasers.

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