Giant photoluminescence enhancement in deuterated highly strained InAs/GaAs quantum wells

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The photoluminescence of InAs/GaAs pseudomorphic single quantum wells, of width 1, 1.2, and 1.6 monolayers, is studied before and after diffusion of monoatomic deuterium into the samples. The luminescence shows a red shift for increasing nominal well width, suggesting an interface roughness on a scale much smaller than the exciton size. The luminescence efficiency increases by several orders of magnitude after sample deuteration. A discussion about the origin of radiative recombination in these heterostructures, before and after deuteration, is also given.

InAs/GaAs highly strained quantum wells have potential applications in electronic and optoelectronic devices. Their pseudomorphic growth is made difficult by the large lattice mismatch (ε=7.16%) between InAs and GaAs. The critical thickness (t_c) for InAs grown on GaAs is indeed less than 3 monolayers (ML). However an island morphology may occur during growth already for layers thinner than t_c, thus introducing defects in the quantum well which severely affect the electronic characteristics, in particular the photoluminescence (PL) efficiency.[1,2]

In this work we study the low temperature photoluminescence of pseudomorphic InAs/GaAs quantum wells before and after deuterium irradiation. Untreated samples show two recombination bands. After deuterium irradiation the whole PL intensity is strongly enhanced, up to three orders of magnitude for the highest diffusion dose. Moreover, the line shape and the energy position of the low-energy recombination are changed. This points out that deuterium (or more generally hydrogen) efficiently passivates nonradiative centers,[3,5] thus making InAs/GaAs quantum wells more attractive for optoelectronic applications.

Three InAs/GaAs single-quantum-well (SQW) structures have been grown by molecular beam epitaxy on GaAs(100) substrates. The nominal thicknesses of the InAs SQWs are 1 ML (sample M1), 1.2 ML (M2), and 1.6 ML (M3). The fractional InAs coverage indicates the percentage of the sample surface covered by the second monolayer. The growth conditions have been set after the work by Brandt and co-workers.[6] A 0.5-μm-thick buffer layer has been grown at 520 °C. The growth temperature has been subsequently lowered to 420 °C before growing the InAs wells. On top of the InAs layers, 10 ML of GaAs have been grown at the same temperature. The growth temperature has then been raised again to 520 °C, to grow a 40-nm-thick GaAs cap layer. The growth rate for InAs was 0.1 ML/s under As-stabilized conditions. During the InAs growth, the reflection high energy electron diffraction has remained that typical for a two-dimensional growth.

Deuterium diffusion was produced by ion-beam irradiation from a Kaufman source of samples held at 300 °C. The ion energy was about 100 eV, the current density of order tens of μA cm^{-2}. Several doses have been used in this study. In the sample name, the letter indicates the implantation dose: ν for the untreated sample, and $\alpha,\beta,\gamma,\delta,\eta$, respectively, for 5.6×10^{14}, 2.8×10^{15}, 2×10^{16}, 1.4×10^{17}, and 1.3×10^{18} ions cm^{-2}. For each dose, all samples have been deuterated in a single run. PL measurements have been performed at several temperatures. The excitation wavelength was 458 nm, when not otherwise mentioned.

Figure 1 shows the PL spectrum of the three untreated samples at 5 K. The doublet observed in sample M1ν (1 ML) is very similar in shape and energy position to those observed in previous works.[7,8] The low-energy, broad band A has been attributed to carbon acceptors in Ref. 7, remaining unidentified in Ref. 8. Its behavior with temperature and power is not that typical for an impurity recombination.[9] Band A becomes broader and smoothly weakens for increasing temperature, being no longer resolved for $T>77$ K. Its intensity relative to that of band B remains almost constant up to 35 K, while its energy separation from band B decreases with increasing temperature. The dependence of the intensity of band A on the excitation power is superlinear and we do not observe the saturation expected for recombination at impurity sites.[10] The most striking effect is the blue shift

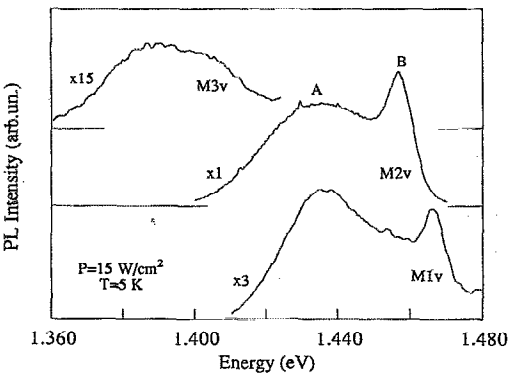

FIG. 1. PL spectra of untreated M1, M2, and M3 samples at T=5 K. Note that the signal intensity is comparable in M1 and M2, while it is one order of magnitude weaker in M3.

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TABLE I. Experimental and theoretical values for the HHFE transition energy. The values used to determine the zero-point energy in a simple square-well model are conduction-band offset $\Delta E_c=0.7\ \Delta E_g$, $m_e=0.067m_0$, $m_{hh}=0.45m_0$. The effective masses are those of GaAs because most of the wavefunction extends into the GaAs barriers. For M3, the experimental value for high excitation intensity is reported.

Sample	HHFE energy (eV) (experiment)	HHFE energy (eV) (theory)
M1 (1 ML)	1.466	1.471
M2 (1.2 ML)	1.457	1.453
M3 (1.6 ML)	1.403	1.413

of band A with increasing excitation. This feature suggests the presence of a continuum of states rather than a single state, as a low concentration of impurity should produce. Recombination from donor-acceptor pairs is also ruled out[10] by the high quenching temperature of band A. Moreover, we wish to point out that an impurity recombination has never been observed in InGaAs/GaAs QWs grown in our laboratory (the residual doping is in the 10^{14} cm^{-3} range).

In agreement with previous works,[7,8] we attribute band B to the recombination of the heavy-hole free exciton (HHFE). Its full width at half-maximum (FWHM) is 6 meV in sample M1 and 5 meV in M2. These values well compare with those previously obtained,[7,8] showing the state-of-the-art quality of the samples. The spectrum of M2 (1.2 ML) is very similar to that of M1, the only differences being a red shift and a smaller energy separation between band A and band B. The spectrum of M3 (1.6 ML), instead, differs from the previous spectra, the recombination from band A being now dominant.

One of the most controversial topics in quantum-well research is the interface roughness, in particular its lateral scale. The shifts to lower energy for wider wells (1.2 and 1.6 ML) in Fig. 1 suggests that the excitons experience an effective well width equal to the nominal one. This is only possible if the InAs islands having 1 or 2 ML thickness are much smaller in size than the exciton diameter. In the opposite case (formation of monolayer-flat islands larger than the exciton diameter), the PL spectrum should always show two free-exciton peaks, corresponding to recombination in 1- and 2-monolayer-thick sample regions. In Table I, we compare the experimental energies with those calculated for the HHFE ground state in a simple square-well model, with a fully strained InAs layer. For sample M3, we have taken 1.403 eV as the experimental peak energy, value obtained at high excitation intensity, where the free-exciton contribution is strongest. The agreement between theory and experiment is quite good. The comparison indicates that in M3 a sizeable strain relaxation with generation of dislocations has not occurred.

As to band A, its feature described above rule our an impurity origin. Moreover, after our calculations about 90% of the HHFE wave function is spread out in the barrier region. The binding energy of an impurity should therefore decrease with decreasing well width, converging to the impurity binding energy in GaAs. On the contrary, by narrowing the well width we observe an increase of the energy distance between bands A and B. We suggest, therefore, a

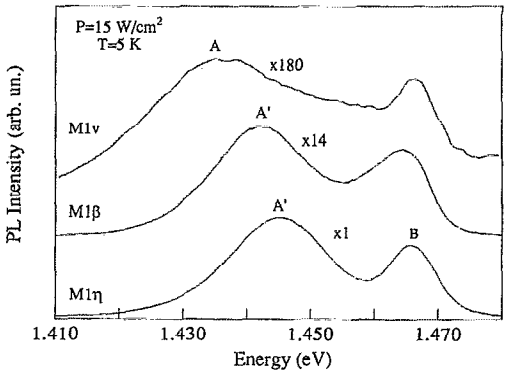

FIG. 2. PL spectra of samples M1ν, M1β (irradiation dose 2.8×10^{15} ions cm^{-2}), and M1η (irradiation dose 1.3×10^{18} ions cm^{-2}) at 5 K.

new process for the origin of band A: it originates from particle localization by interface defects or by the potential fluctuations associated with the thickness variation around the mean, nominal well-width value. Our attribution is supported by the observation that, while the spectra of M1 and M2 are very similar, except for the energy shift, the spectrum of M3 corresponds to a comparatively stronger low-energy recombination, as suggested by the broad linewidth and by its temperature dependence. The quantum well having 1.6 ML thickness, although does not show a sizeable strain relaxation, probably presents the highest degree of interface disorder among the three samples, and thus the strongest localization effects.

We now come to the effect of introducing deuterium into the samples. In Fig. 2, we show the PL spectra of samples M1ν, M1β, M1η. Deuterium incorporation causes a strong increase (more than hundred times for M1 and M2, about one thousand for M3) of the integrated luminescence intensity. The ratio (I_D/I_V) between the intensities of the deuterated samples and that of the untreated samples is reported as a function of the irradiation dose in Fig. 3, for $T=5$ K. No major differences are observed at higher temperatures of measurement. The PL enhancement after deuteration is a smooth function of excitation intensity, decreasing at high

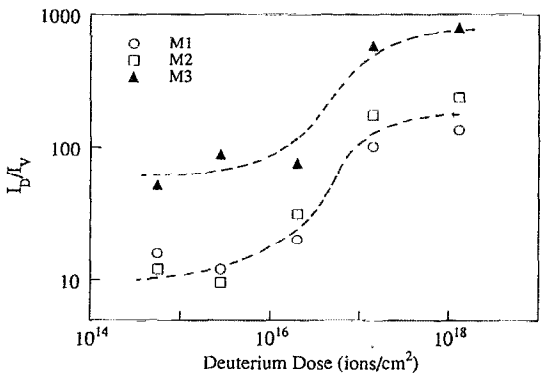

FIG. 3. Ratio of the PL integrated intensity between deuterated and untreated samples as a function of the irradiation dose. The excitation intensity was 15 W cm^{-2}, the temperature 5 K. Dashed lines are guides to the eye.

excitation intensity. The similarity of the luminescence characteristics of samples M1v and M2v is reflected in a similar enhancement curve with the deuterium dose, which seems to saturate at the highest doses. The stronger enhancement observed for M3 is likely to be related again to the presence of a larger number of interface defects, playing a role in nonradiative recombination. Indeed, in the untreated samples, M3 has an integrated PL intensity one order of magnitude weaker than M1 and M2, while, in deuterated samples, the PL intensities become comparable.

In addition to the strong efficiency enhancement of the overall luminescence, we observe some change in the luminescence line shape. At low energy, a broad emission is observed (band A′) which is narrower and blue shifted with respect to band A in the untreated sample. The blue shift increases with the deuterium dose. A similar behavior is observed in deuterated M2 and M3 samples. The presence of band A′ (as well as of band A) in spectra taken with an excitation wavelength of 830 nm (i.e., below the GaAs band gap) confirms that it originates in the InAs layer.

It is not easy to ascertain if band A′ in deuterated samples has the same origin as band A in untreated samples. In InGaAs/GaAs quantum wells, a broad, intense emission occurs in deuterated samples, 15–25 meV below the HHFE emission, depending on the well thickness.[11,12] Contrary to the present InAs/GaAs case, no emission has been observed at the same energy in the untreated InGaAs/GaAs samples. In Ref. 11, the broad band in deuterated samples has been attributed to an impurity-like recombination at a deuterium-defect complex (most likely at the interface). A similar picture could apply to band A′ in the present work. In any case, both band A′ and band A lose importance at high temperatures and excitation intensities.

In conclusion, we have analyzed the photoluminescence of untreated and deuterated InAs/GaAs SQWs with thickness around 1 ML. We have attributed the low-energy recombination observed in untreated samples to particle localization at interface defects or atomic-scale roughness. The giant increase of the luminescence efficiency in deuterated samples shows that deuterium incorporation has a beneficial effect on these highly strained heterostructures, making them more suitable for optoelectronic applications.

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