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Evidence for hydrogen accumulation at strained layer heterojunctions

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The incorporation of hydrogen into strained $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ quantum wells results in the formation of shallow, H-related radiative states which compete with, and quench, the intrinsic band-to-band luminescence. By comparing the photoluminescence data obtained from hydrogenated material with secondary ion mass spectroscopy profiles from deuterated material, it is possible to deduce that the H-related radiative states are associated with H which is accumulated at the well interfaces.

Many of the experimental studies relating to the introduction of H into III-V semiconductors reflect the fact that the central role of passivation is achieved in much the same way as for Si. However, distinct differences are observed between lattice-matched GaAs/AlGaAs and strained InGaAs/GaAs heterojunctions, which have been hydrogenated in a similar manner. The principal effect of incorporating H into GaAs/AlGaAs is to decrease the number of nonradiative centers available for recombination, thereby increasing the photoluminescence (PL) efficiency. Above an optimal H dose, additional radiative states become both activated and created due to the very strong but local bond distortion wrought by H to the GaAs lattice.¹ To date, such increases, in both PL intensity and carrier lifetime, have been observed for single GaAs/AlGaAs heterojunctions,¹ short period GaAs/AlAs superlattices,² InGaAs/AlGaAs quantum wells (QWs),³ and near-surface GaAs/AlGaAs (Ref. 4) QWs. The consensus of opinion indicates that hydrogen predominantly passivates interface defects in all GaAs/Al_xGa_{1-x}As heterojunctions.

In contrast, low temperature PL measurements, following the post-growth hydrogenation of InGaAs/GaAs QWs, have revealed^{5,6} the formation of shallow, H-related states, at even low H doses, which compete with and quench the intrinsic band-to-band recombination. Furthermore, the sharp decrease in intensity of these H-related transitions, with increasing distance of the QW from the sample surface, has led to the speculation that the H profile, within these crystals, may behave in a similar manner. To some extent, this apparent inhibition has been verified by hydrogenating similar QW structures, which differed⁷ only in In alloy composition. PL measurements performed on samples possessing In mole fractions of $x=0.09$, $x=0.16$, and $x=0.20$, revealed that for both $x=0.16$ and $x=0.20$ compositions, shallow H-related radiative recombination arose only from the QW closest to the sample surface. Likewise, hydrogenation⁴ of a structure consisting of 60 and 80 Å $\text{In}_{0.13}\text{Ga}_{0.87}\text{As}/\text{GaAs}$ wells, separated by 3500 Å GaAs, has shown that for H doses up to 10^{18} cm^{-2} , implanted at 250 °C, H-related PL is only observed from

the 80 Å near-surface QW. However, it is only possible to speak in terms of there being an *inhibition* to the diffusion of H, if the H-related states are deemed to be distributed uniformly throughout each QW region. Hence, the objective of the present letter is to determine if the H-related radiative states are indicative of H bonded within an $\text{In}_x\text{Ga}_{1-x}\text{As}$ environment, or whether this luminescence arises solely from H trapped at the QW interfaces. Optical data obtained from hydrogenated InGaAs/GaAs QW structures are outlined first, before proceeding to discuss the secondary ion mass spectroscopy (SIMS) profiles obtained for deuterated material. The combination of PL and SIMS data provide evidence to suggest that the H-related radiative states, present in the PL measurements, reflect the excess H which is accumulated at the strained QW interfaces.

The strained, undoped $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ QW samples used in this study were grown at 500 °C, by molecular beam epitaxy (MBE), inside a VG Semicon V80H MBE reactor. Hydrogenation was carried out with a Kauffmann ion source, operated at 100 eV, while the sample temperature was maintained at 300 °C. The accelerating voltage was increased to 200 eV for D implantation. An ion current of $100 \mu\text{A}/\text{cm}^2$ was used in both cases. The samples were analyzed with an Atomika SIMS system employing normal incidence oxygen bombardment ($1 \mu\text{A}$ at 12 keV). The SIMS raster size was $600 \times 600 \mu\text{m}$. For calibration purposes, a standard H implant in GaAs was also analyzed. It has been assumed that the system response for both H and D is identical; this assumption being valid to within a factor of 2, and it follows that the accuracy of the concentration measurements (for D) bears a similar uncertainty. The thickness of the outermost well (160 Å) has been used for depth calibration purposes. The samples chosen for deuteration consisted of 20, 40, 80, and 160 Å $\text{In}_{0.20}\text{Ga}_{0.80}\text{As}/\text{GaAs}$ wells (growth order) separated by 1000 Å GaAs barriers.

Figure 1(a) shows the 4.2 K PL spectrum obtained from an $\text{In}_{0.09}\text{Ga}_{0.91}\text{As}/\text{GaAs}$ QW sample, hydrogenated with $2 \times 10^{18} \text{ H ions}/\text{cm}^2$. The structure consists of 20, 40, 80, and 160 Å QWs, separated by 500 Å GaAs barriers, and with a 500 Å GaAs capping layer. Hence, the lowest energy *intrinsic* PL transition arises from the QW which is

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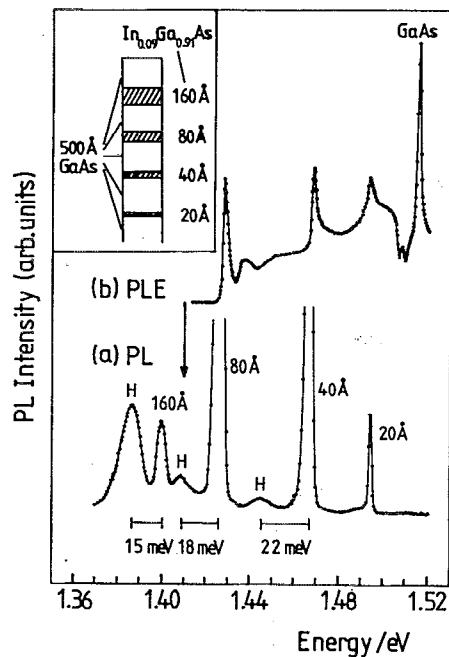


FIG. 1. (a) 4.2 K PL spectrum obtained from the $\text{In}_{0.09}\text{Ga}_{0.91}\text{As}/\text{GaAs}$ QW structure shown in the inset, hydrogenated with 2×10^{18} H ions/ cm^2 , and (b) 4.2 K PLE spectrum for the H-related PL band arising from the 80 Å QW.

closest to the surface of the sample. Implantation of H ions results in three additional, shallow H-related PL peaks, each of which is a satellite to the *intrinsic* transitions within the 160, 80, and 40 Å QWs, respectively. The intensities of these H-related PL features decrease rapidly, with increasing distance from the sample surface. Figure 1(b) is the PL excitation (PLE) spectrum obtained by monitoring the PL intensity at 1.410 eV, the peak of the H-related recombination for the 80 Å QW. The onset of excitation, 1.428 eV, corresponds to the $e_1\text{-}hh_1$ transition within the 80 Å well, thus giving a value of 18 ± 0.5 meV for the binding energy of this H-related transition. The next step in the PLE spectrum corresponds to the onset of continuum excitation for the 80 Å QW, and so the energy difference between these two PLE features provides a value for the exciton binding energy of 8.0 ± 0.5 meV, in this case. Further peaks occur at higher energies in the PLE spectrum (but still below the band gap of the GaAs barrier material), which reflect *indirect* excitation of radiative transitions within the 80 Å QW, via *photon recycling* from the 40 and 20 Å wells. Both the line shape of the PL spectrum and the quality of the PLE spectrum indicate that the H-related luminescence cannot be attributed to bombardment damage. Furthermore, hydrogenation of near-surface GaAs/AlGaAs QWs, in a similar manner,⁴ has shown an actual increase in the normalized QW PL intensity, after hydrogenation, thus revealing passivation of interfacial nonradiative recombination centers for structures with AlGaAs surface barriers below 120 Å in thickness. In the present case, the observed⁵ variation in binding energy, from 22 meV (40 Å QW) to 15 meV (160 Å QW) for the H-related states within the $\text{In}_{0.09}\text{Ga}_{0.91}\text{As}/\text{GaAs}$ QWs, is fully consistent

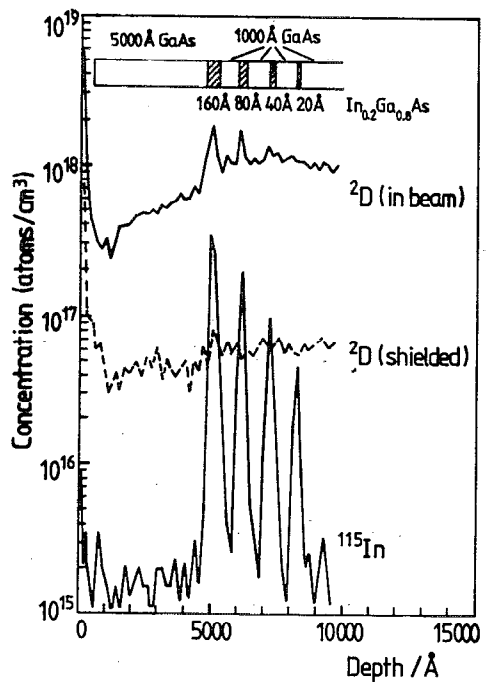


FIG. 2. ^2D and ^{115}In SIMS profiles obtained from an $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ QW sample, deuterated with 5×10^{18} ions/ cm^2 . The ^2D (shielded) trace reveals the background detection limit for a region of sample masked from the D beam.

with that predicted⁸ for quantized shallow states. Since the structure was grown without intentional doping, it is lightly *p* type, and hence hydrogenation results predominantly in the formation of donorlike centers. At the same time, the magnitude of the binding energy suggests that the H-related states are *effective-mass* defects, which arise from the strong local bond distortion associated with the introduction of hydrogen.

Figure 2 contains calibrated SIMS profiles obtained for ^2D (in-beam and shielded regions), plus ^{115}In (in beam only), for the sample with a 5000 Å GaAs cap layer, deuterated with 5×10^{18} impinging ions/ cm^2 . The apparent D concentration of 5×10^{16} cm^{-3} , obtained in the region shielded from the D beam, represents the background limit of the system due to the presence of molecular hydrogen (indistinguishable from D). An arbitrary concentration factor has been applied to the In profile, which has also been displaced vertically in the figure for clarity. The profile obtained from the deuterated area reveals that the D penetrates the whole of the structure, with a steady-state concentration of about 1×10^{18} cm^{-3} . However, D spikes are also observed, which correlate with the positions of the In peaks obtained from each of the QWs. These spikes are clearly not due to selective adsorption of molecular H during analysis, since they are not present in the D profile obtained from the shielded region. The intensities of these spikes decrease with increasing distance from the sample surface, making it difficult to discern a clear spike which correlates with the location of the 20 Å QW.

Figure 3 depicts ^2D and ^{115}In profiles obtained from the sample with a 2000 Å GaAs cap layer. Once again, there is a homogeneous level of about 10^{18} cm^{-3} D set,

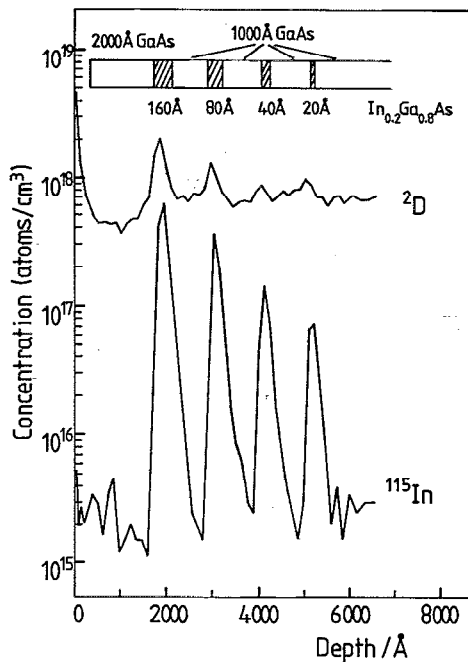


FIG. 3. ^2D and ^{115}In SIMS profiles obtained from an $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ QW sample, deuterated with 5×10^{18} ions/cm 2 .

presumably at such high dosage (5×10^{18} impinging ions/cm 2) by the total density of D-trapping defects. In this case, a distinct D peak is evident for all four QWs in the structure. For comparison, SIMS data obtained³ from the $\text{InGaAs}/\text{AlGaAs}$ system has shown distinct D peaks, located in the vicinity of the well interfaces, for relaxed 400 and 800 Å QWs. However, in that case it was not possible to resolve the D signal between the thinner strained wells and their corresponding barriers. If the D spikes correspond to D distributed uniformly throughout each QW, then the amplitude of each spike should be proportional to the well width. Since the well width varies from 160 to 20 Å, with increasing distance from the sample surface, then the ratio in spike amplitudes (D_{160}/D_{20}) should be ≥ 8.0 . On the other hand, if the D spikes reflect D which is accumulated only at the well interfaces, then variations in spike amplitude should follow directly the excess amount of interfacial D, above any continuous background. The deuterated sample with a 2000 Å GaAs surface barrier has $D_{160}/D_{20} = 3.4$, significantly below that required for a uniform distribution of D throughout each QW. The increased value for $D_{160}/D_{20} \geq 4.6$ from the sample with a 5000 Å GaAs cap layer is consistent with observations⁴ for the relative intensity of the H-related PL peak; increasing the thickness of the GaAs surface barrier has the same effect as decreasing the H dose.

To summarize, the variation in intensity of the D

spikes, with increasing distance from the sample surface, mirrors the trend observed in Fig. 1 for the H-related PL peak intensities. This leads to the proposal that the H-related radiative recombination centers are associated with an accumulation of H, in excess of the uniform volume concentrations, at the $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ QW interfaces. Since similar shallow, H-related PL bands have not been observed⁴ to date, following hydrogenation of GaAs/AlGaAs QW structures, it seems that these H-induced radiative states are characteristic of the strained $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ heterojunction. Both SIM⁹ and photoemission¹⁰ measurements, coupled with PL data, have revealed recently that there exists a variable degree of intermixing at the $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ interface due to the surface segregation of In atoms during growth. Hence, at present, it remains difficult to speculate about the relationship between the H-related recombination centers and the atomic configuration of the interfacially bonded hydrogen atoms. In conclusion, a combination of PL and SIMS data provide evidence to suggest that H-related radiative states, which arise in PL measurements for hydrogenated $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ QWs, are associated with H which is accumulated at the strained QW interfaces.

The material used throughout these studies was grown by David Woolf and David Westwood, at U.W.C.C. Hydrogenation was carried out in Dipartimento di Fisica, Università di Roma, in collaboration with Mario Capizzi, Carlo Coluzza, and Andrea Frova. PLE measurements were in association (Ref. 5) with Richard Phillips at the University of Exeter. The financial assistance of the U. K. Science and Engineering Research Council (SERC) is gratefully acknowledged, under the Low-Dimensional Structures and Devices Initiative.

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