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Layer perfection in ultrathin MOVPE-grown InAs layers buried in GaAs(001) studied by X-ray standing waves and photoluminescence spectroscopy

James A. Gupta,[a] Joseph C. Woicik,[b] Simon P. Watkins,[a]* Dale A. Harrison,[a] E. Daryl Crozier[a] and Barry A. Karlin[b]

[a]Department of Physics, Simon Fraser University, Burnaby, B.C. V5A 1S6 Canada, and [b]National Institute of Standards and Technology, Gaithersburg, Maryland, 20899 USA. E-mail: simonw@sfu.ca

Using the In-L fluorescence produced by normal-incidence X-ray standing waves, we have measured the layer perfection and positions of 1 and 1/2 monolayer (ML) InAs quantum wells buried in GaAs(001). Growth temperature effects were studied in a series of samples produced by metalorganic vapor phase epitaxy (MOVPE) at temperatures between 400 and 600°C. The coherent position of the In atoms decreases with temperature in the 1 ML samples, and the optimal growth temperature is near 550°C, as evidenced by the coherent position of 1.15±0.02 and the relatively high coherent fraction of 0.72±0.08. These results are corroborated by 1.6 K photoluminescence (PL) measurements in which the most sharp and intense In-excitonic emission is obtained from a sample grown at 530°C. For the 1/2 ML samples, growth temperatures of 400°C and 600°C produce similar standing wave results: coherent positions of 1.09±0.02 and 1.10±0.02, coherent fractions of 0.75±0.10 and 0.74±0.11, respectively. However, PL reveals the higher temperature sample to be of far superior quality, due to excessive carbon incorporation at 400°C.

Keywords: X-Ray standing waves; photoluminescence spectroscopy; InAs quantum wells.

1. Introduction

Buried films of InAs in GaAs(001) constitute one of the most highly-strained III-V semiconductor heterostructures. The ~7% lattice mismatch results in a critical thickness for coherent epitaxy of only ~1.5 MLs, making this system ideal for the study of fundamental materials issues [Giannini *et al.* (1993), Woicik *et al.* (1995), Lee *et al* (1996)]. Additionally, very recent theoretical [Iotti *et al.* (1998)] and experimental [Goñi *et al.* (1998)] studies have re-emphasized the technological promise of InAs single quantum wells for highly efficient excitonic lasing. Here we use back-reflection X-ray standing waves (XSW) to obtain the structure of a series of MOVPE-grown films in order to characterize the In atomic distribution and provide a structural basis for understanding the optical (PL) properties of the material. In the back- reflection mode the incident photon energy is scanned through the Bragg condition at normal incidence. As such, the technique may be used on synchrotron spectroscopy beamlines to complement XAFS measurements. Notably, the In atomic signature is measured via the emission of fluorescent X-ray photons, where the incident energies lie above an absorption edge.

2. Experiment

Epitaxy was performed in a vertical MOVPE reactor on semi-insulating GaAs(001) substrates. Following annealing under tertiarybutylarsine (TBAs) at 580 °C, GaAs buffer layers were grown using triethylgallium (TEGa) and TBAs. Each sample was then allowed to equilibrate at the desired growth temperature, T_g. GaAs barrier layers of thickness ~100 Å were deposited above and below the InAs layer which was also grown at T_g using trimethylindium and TBAs. For the PL samples an additional GaAs cap layer (~1900 Å) was deposited at 580°C.

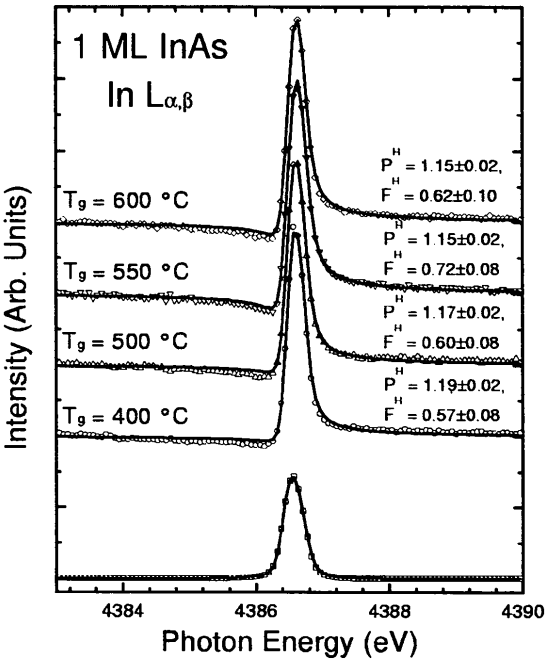

Figure 1
Photon-energy dependence of the reflectivity and the In-L fluorescence yield near the GaAs(004) Bragg backreflection condition for a series of 1 ML InAs samples, with growth temperatures as indicated. The solid lines are the best fits to the data points. The spectra were offset for clarity.

Standing waves were produced within the samples and modulated in phase by scanning in energy through the GaAs (004) back-reflection at normal incidence. Fluorescence yield data were obtained using a single-element Ge(Li) detector in the same manner as for fluorescence XAFS experiments. Reflectivity measurements were made by recording the back-reflected photon intensity with a Ni mesh upstream from the sample.

Low-temperature photoluminescence measurements were performed at 1.6 K using Ar-ion laser excitation. Spectra were recorded with a Fourier transform interferometer.

3. Results and Discussion

X-ray reflectivities and the resulting X-ray standing wave patterns were recorded simultaneously [Zegenhagen (1993)] as shown in

Figures 1 and 2. The standing wave results from the superposition of the incident and back-reflected travelling waves and has the periodicity of the crystal lattice planes with diffraction vector, $\vec{H}$. For energies below the Bragg reflection the nodes of the standing wave lie on the substrate planes. As the phase shifts throughout the Bragg peak the wavefield moves continuously until the antinodes lie on the substrate planes at the high energy side. Indium fluorescence spectra (Figures 1 and 2) result from the absorption of the In atoms in the field of the standing wave. From this response the spatial distribution of the In atoms may be deduced.

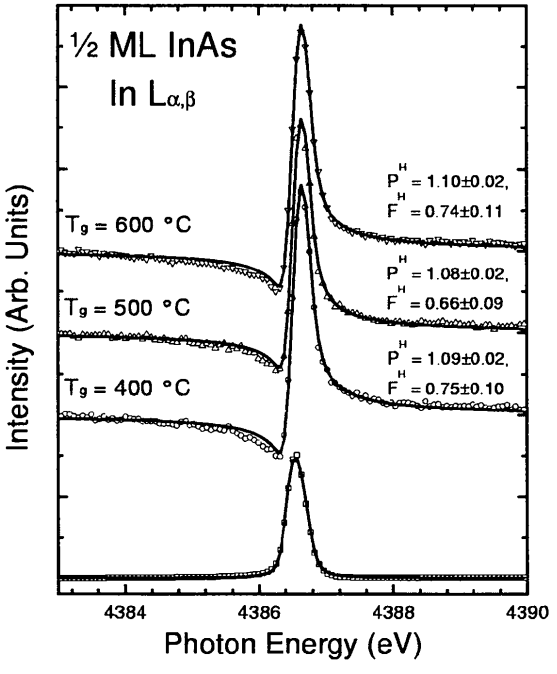

Figure 2
Photon-energy dependence of the reflectivity and the In-L fluorescence yield near the GaAs(004) Bragg backreflection condition for a series of 1/2 ML InAs samples, with growth temperatures as indicated. The solid lines are the best fits to the data points. The spectra were offset for clarity.

The yield from the standing wave measurement is written [Zegenhagen (1993)] in terms of the coherent fraction, F^H, which measures the δ-like nature of the atomic distribution, and the coherent position, P^H, which measures the position of the In atoms relative to the substrate planes, normalized by the diffraction plane spacing, d_s. (For a single atom in bulk P^H=1). The yield, Y^H, is

$$Y^H = Y^0(1 + R + 2\sqrt{R}F^H cos(\alpha - 2\pi P^H)) \ . \qquad (1)$$

Here R and α denote the reflectivity and phase as given by the dynamical theory of X-ray diffraction [Zegenhagen (1993)], and Y^0 is proportional to the beam flux, depending linearly on energy. Thermal vibrations are taken into account by a Debye-Waller factor which decreases the measured value of F^H.

From the macroscopic elastic theory [Hornstra & Bartels (1978)] , a single InAs ML in GaAs is expected to have P^H = 1.146 under pseudomorphic conditions. Our best sample, structurally, is the one with T_g = 550°C, having an ideal coherent position and the highest F^H. 1/2 ML samples grown at 400 and 600°C have similar positional values to one another and very high values of F^H.

Recently [Gupta *et al.* (1998)], we used a simple macroscopic elastic theory calculation to interpret results for nominally 1 and 1/2 ML samples grown by atomic layer epitaxy. Using Vegard's law and the elastic constants of InAs and GaAs it was shown that the XSW position may be related to the composition of $In_xGa_{1-x}As$ layers, for less than one full ML of InAs. Using this scheme, we find that the present 1/2 ML samples have In alloy compositions, x, of 0.63±0.14, 0.56±0.14 and 0.69±0.14, in order of increasing T_g.

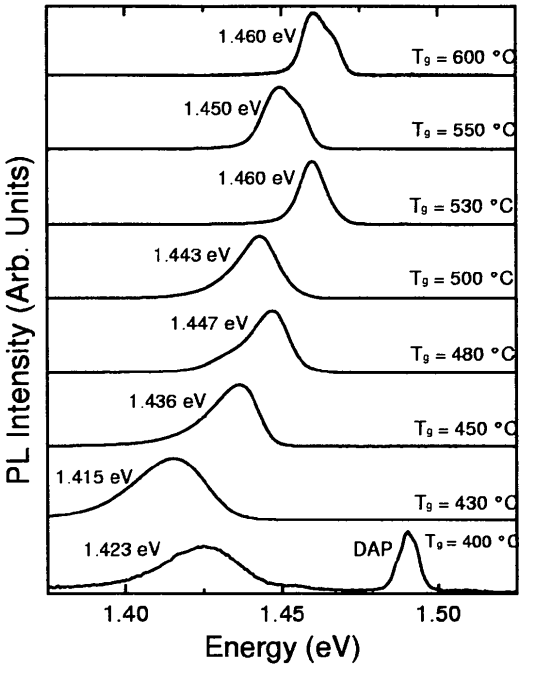

Figure 3
Photoluminescence spectra for a series of 1 ML InAs samples, with growth temperatures as indicated. The spectra were normalized to a peak height of unity and offset for clarity.

The PL spectrum (Figure 3) for the sample grown at 400°C shows very weak In-excitonic luminescence. The features labeled 'DAP' represent donor-acceptor pair (1.490 eV) and free-to-bound (1.493 eV) transitions at carbon impurities. These result from the incomplete decomposition of TBAs precursor molecules, leading to carbon incorporation in the crystal. For the remaining samples, the PL emission energies increase with growth temperature, while the linewidths generally decrease. An exception is the sample with T_g = 530°C, exhibiting particularly narrow and bright luminescence.

For the 1/2 ML samples (Figure 4), 400°C is again seen to be unsuitable for good optical material, despite the more promising

XSW results. The spectrum is dominated by the carbon impurity features. The brightest 1/2 ML In-luminescence is observed for the sample with $T_g = 600°C$.

A comparison between the structural and optical properties appears in Figure 5. The data suggest a correspondence between the decreasing coherent position and the increasing PL peak energies as a function of growth temperature in the 1 ML samples. As well, the PL linewidths decrease dramatically with increasing growth temperature. This is mirrored somewhat by the increasing coherent fraction. These characteristics may be attributed to a trend towards more perfect, δ-like InAs layers, being particularly evident in the 530-550°C range.

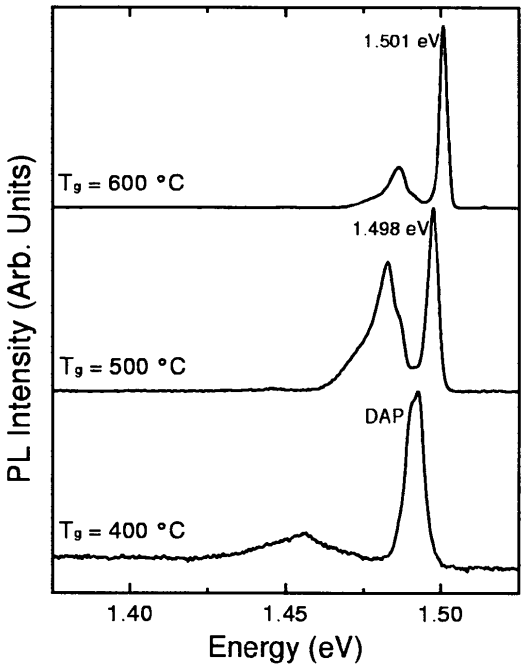

Figure 4
Photoluminescence spectra for a series of 1/2 ML InAs samples, with growth temperatures as indicated. The spectra were normalized to a peak height of unity and offset for clarity.

The lack of any clear trend for the 1/2 ML samples is noteworthy; since the MOVPE InAs growth exposures for these samples were half the corresponding values for the full MLs, we might expect similarities between the two series. The lack thereof may be related to details of the growth mechanism at the subML scale. For example, 2-dimensional versus 3-dimensional modes, or issues of segregation and compositional alloying. An adequate interpretation of this data will benefit from direct measurements of the strain and bond configurations using X-ray absorption spectroscopy.

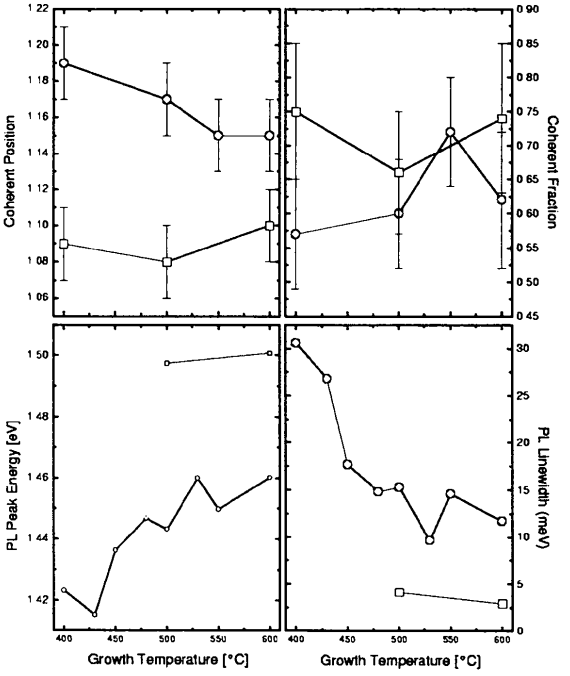

Figure 5
Summary of the XSW and PL results for all samples. The circles and squares correspond to 1 ML and 1/2 ML samples, respectively. For the PL data, the uncertainties are given approximately by the symbol size. The solid lines are guides for the eye.

In conclusion, we have used the XSW technique to assess the structural perfection of monolayer and submonolayer InAs quantum wells in GaAs. Our measurements show morphological differences in samples grown at different temperatures, and yield a quantitative measure of the interface perfection. Accompanied by PL measurements, this allows us to evaluate the growth conditions to optimize the quality of the material for light-emitting devices.

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