InGaP/GaAs superlattices grown by gas-source molecular beam epitaxy


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Lattice-matched InGaP/GaAs superlattices have been grown by gas-source molecular beam epitaxy. High-resolution images obtained with transmission electron microscopy reveal the superlattices to be free of dislocations and to exhibit smooth interfaces of only 1–2 monolayers in width. Double crystal x-ray diffraction studies indicate that the narrow interfacial regions are locally strained as a result of the growth sequence during gas-source molecular beam epitaxy.

The materials system InGaP/GaAs offers an alternative to AlGaAs/GaAs for fabrication of multiple quantum well (MQW) optoelectronic devices. The valence-band offset energy is larger for the InGaP/GaAs interface than for the AlGaAs/GaAs interface, and InGaP when lattice matched to GaAs, exhibits a much lower concentration of deep levels than AlGaAs.1 Superlattices of AlGaAs/GaAs have been extensively characterized and MQWs with atomically abrupt interfaces have been obtained with layers grown by molecular beam epitaxy (MBE) and metalorganic chemical vapor deposition (MOCVD). However, comparatively little is known about InGaP/GaAs superlattices. Razeghi et al. have recently reported InGaP/GaAs superlattices grown by low-pressure MOCVD.2 We present here the results of a detailed study of nominally lattice-matched InGaP/GaAs superlattices grown by gas-source MBE. The InGaP/GaAs heterostructures are found to exhibit smooth interfaces with an interfacial roughness of 1–2 monolayers but with interfacial strain apparently resulting from the growth sequence during gas-source MBE.

Two superlattice samples were grown on (100) GaAs substrates at approximately 520 °C using conventional Ga and In MBE effusion cells and As5 and P2 molecular beams produced by thermal decomposition of AsH3 and PH3, respectively, in a low-pressure cracking oven held at 900 °C. The InGaP layers were grown under conditions similar to that previously reported3 with a growth rate of 0.5 µm/h and PH3 flow rate of 5 sccm. During the GaAs layer grown at 0.3 µm/h, the AsH3 flow rate was 2 sccm.

The use of the gas sources provided a practical method of controlling the group V molecular flux while switching between InGaP and GaAs growth.4 At each interface the growth was temporarily stopped by first closing the group III shutters, then after 6 s the As5 and P2 beams were switched, and an additional 18 s the growth was resumed by opening the appropriate group III shutters. This same growth sequence has been used to produce abrupt InGaP/GaAs heterointerfaces in single quantum wells as narrow as 0.6 nm.5

The layer thicknesses and interface quality of the InGaP/GaAs superlattice samples were determined by transmission electron microscopy (TEM) using a JEM 2000 EX microscope operated at 200 kV. Figure 1 is a (200) dark field image of a [011] cross section of a sample consisting of a 100 nm GaAs buffer layer, a 612 nm InGaP buffer layer, and a 20-period superlattice with each period containing a GaAs well layer (dark regions in Fig. 1) and an InGaP barrier layer (bright regions), all covered with a 50 nm GaAs cap layer. The superlattice layers are uniform in thickness with smooth distinct interfaces. The cross section of Fig. 1 compares favorably with the TEM images at a similar magnification of InGaP/GaAs superlattices grown on GaAs by MOCVD.2

Figure 2 is a high-resolution TEM image of a [011] cross section of a GaAs well layer in the sample of Fig. 1. Our JEM microscope is equipped with an ultrahigh resolution pole piece which at the operating voltage yields a point resolution of 0.20 nm, and the defocusing for the image in Fig. 2 is close to the Scherzer focus of the microscope. The width of the heterointerfaces is seen to be 1–2 monolayers, and the structures are found to be free of misfit dislocations, indica-

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tive of coherent growth. The thickness of the GaAs well layer was determined to be $5.9 \pm 0.6$ nm and the InGaP barrier layer to be $25.0 \pm 0.8$ nm, in good agreement with that expected from the growth conditions.

InGaP/GaAs superlattices were also examined by double crystal x-ray (DCX) diffraction using Cu $K_a$ radiation and the (400) reflection. Figure 3(a) is the DCX spectrum for the sample of Fig. 1; the large number of sharp, well-defined satellite peaks is usually interpreted as characteristic of a superlattice of very high structural quality. Using the superlattice peak at $n = 0$ in Fig. 3(a), the relaxed mismatch relative to the GaAs substrate was found to be $0.9 \times 10^{-4}$ and the In$_x$Ga$_{1-x}$P alloy composition $x$ to be 0.483. The superlattice period averaged over the eight

![DCX spectrum of superlattice](image)

FIG. 3. DCX spectrum of a 20-period InGaP/GaAs superlattice with a 5.8 nm GaAs well layer and a 23.7 nm In$_{0.48}$Ga$_{0.52}$P barrier layer. (a) Experimental data for sample of Fig. 1. (b) Stimulated spectrum using a two-layer model without interfacial strain. (c) Stimulated spectrum using a four-layer model that includes interfacial strain.


strongest satellite peaks was $29.5 \pm 0.4$ nm, in agreement with the TEM results. DCX spectra were also simulated by solving the Takagi-Taupin equations$^6$ for the dynamical theory of x-ray scattering. Figure 3(b) shows the simulated spectrum using a two-layer model (i.e., 5.8 nm of GaAs and 23.7 nm of In$_{0.48}$Ga$_{0.52}$P) for each period and assuming atomically abrupt, strain-free interfaces. The simulated spectrum of Fig. 3(b) is a poor fit to the experimental data, with the intensities of the satellite peaks orders of magnitude smaller than measured. No physically reasonable combination of layer thickness and InGaP composition using the two-layer model could be found to produce an adequate fit to the experimental data. However, when a four-layer model which included strained interfacial layers of 1–2 monolayers in thickness was used, good agreement was obtained. A systematic examination of over 200 simulations revealed the intensity of the satellite peaks to be very sensitive to the thickness and composition of the interfacial regions, and the best fit was obtained when intermixing of only the group V sublattice was assumed to occur at each interface. Figure 3(c) is the best-matched simulated spectra where the four-layer model consisted of 5.2 nm of GaAs, 0.6 nm of GaP, 23.1 nm of In$_{0.48}$Ga$_{0.52}$P, and 0.6 nm of In$_{0.48}$Ga$_{0.52}$As$_{0.50}$P$_{0.40}$. The two monolayers of GaP place the GaAs-to-InGaP interface in tension and the two monolayers of In$_{0.48}$Ga$_{0.52}$As$_{0.50}$P$_{0.40}$ place the InGaP-to-GaAs interface in compression.

In an effort to test the validity of the four-layer model, a second InGaP/GaAs sample with a superlattice containing 30 periods of 8.8 nm of GaAs and 12.4 nm of In$_{0.48}$Ga$_{0.52}$P was examined by DCX diffraction. The superlattice in the previous sample was in tension; this superlattice is in compression with a relaxed mismatch of $6.7 \times 10^{-4}$. Figure 4(a) is the experimental spectrum and Fig. 4(b) is the simulated spectrum using the two-layer model. Figure 4(c) was obtained using a four-layer model including two monolayers of GaP at the GaAs-to-InGaP interface and two monolayers of In$_{0.48}$Ga$_{0.52}$As$_{0.50}$P$_{0.40}$ at the InGaP-to-GaAs interface, the same interfacial layer compositions and thicknesses used to obtain Fig. 3(c) for the first sample.

The interfacial compositions in the four-layer model used to obtain Figs. 3(c) and 4(c) suggest that during the growth pause at each interface a significant exchange of As and P occurs on the sample surface. Little evidence for similar group V exchange has been previously reported. Our growth temperature is above the estimated congruent sublimation temperature$^7$ of $-500^\circ$C for In$_{0.48}$Ga$_{0.52}$AsP and thus some P may have been lost and replaced by As in the InGaP- to-GaAs interface. Davies$^8$ found limited conversion of an InP surface to InAsP at 505$^\circ$C in an As$_4$ beam. The loss of As and replacement of P on the GaAs surface appears unlikely under our growth conditions, although conversion of In$_{0.47}$Ga$_{0.53}$As to In$_{0.47}$Ga$_{0.53}$As$_{0.43}$P$_{0.57}$ has been proposed to explain DCX spectra for gas-source MBE structures.$^9$

An alternative explanation for the strained layers may be the asymmetric ordering of the atomic layers at each interface, as recently reported by Vandenbarg $et al.$ in GaInAs/InP superlattices.$^{10}$ The asymmetric ordering would be produced even at monolayer-abrupt interfaces be-
cause the GaAs growth is terminated in an As layer while the InGaP growth is terminated in a P layer, resulting in two asymmetrical interfaces with equal and opposite strain. A separate DCXR simulation using a four-layer model based on the asymmetric ordering mechanism was performed and a spectrum that almost matched the experimental data as well as Fig. 3(c) was obtained using one monolayer of In$_{0.41}$Ga$_{0.59}$As$_{0.5}$P$_{0.5}$ at the GaAs-to-InGaP interface and one monolayer of GaAs$_{0.5}$P$_{0.5}$ at the InGaP-to-GaAs interface. A similar result was obtained when using the asymmetric ordering in simulation of the second sample. Therefore, at this time it appears that both the group V exchange mechanism and the asymmetric ordering mechanism can account for the observed DCXR spectra.

It is interesting to note that the lack of a large number of satellite peaks in a DCXR rocking curve is usually interpreted as indicative of a superlattice of poor quality. However, we have shown that for InGaP/GaAs superlattices the existence of strained interfacial regions will produce a large number of intense satellite peaks. Furthermore, it should be noted that the strained interfacial regions can significantly alter the angular separation between the zeroth-order superlattice peak and the substrate peak, and thus lead to large errors in calculation of the superlattice mismatch and alloy composition if the interfacial strain is overlooked. This is particularly evident when comparing Figs. 4(a) and 4(b).

In conclusion, we have demonstrated that InGaP/GaAs superlattices when grown by gas-source MBE are free of dislocations and exhibit smooth interfaces which are only 1–2 monolayers in width. Based on modeling of DCXR diffraction spectra, the narrow interfacial regions are locally strained either as a result of the exchange of As and P on the surface of the sample during growth interruption at each interface, or due to asymmetric ordering of the atomic layers intrinsic to each interface.

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