Structural characterization of low-temperature molecular beam epitaxial In$_{0.52}$Al$_{0.48}$As/InP heterolayers

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A systematic study of the structural quality and arsenic content of as-grown In$_{0.52}$Al$_{0.48}$As/InP layers deposited on InP by molecular beam epitaxy at temperatures between 150 and 450 °C was performed using transmission electron microscopy and particle-induced x-ray emission. We found that the amount of As incorporated in the layers generally increases with decreasing growth temperature, with the crystalline quality of the layers being good at growth temperatures higher than 200 °C. At 150 °C, a large density of pyramidal defects is formed, the defects are related to the very large amount of excess As incorporated into the layer. The mechanisms leading to the formation of these defects are discussed. At 200 °C, however, the amount of excess As is lower than expected, and wavy streaks of diffuse scattering are seen in electron diffraction. It is shown that small ordered domains of the CuPt type on the group III atoms are responsible for these features.

In$_{0.52}$Al$_{0.48}$As/InP grown by molecular beam epitaxy (MBE) at ~520 °C is lattice matched to InP and is a material of high resistivity ($p > 10^4 \Omega$ cm). This material becomes even more resistive when the growth temperature is lowered. InAlAs layers grown under low-temperature conditions have been used recently as buffer layers for field-effect transistors, resulting in improved device performance.[1] In a previous study,[2] it was shown that recombination times as low as 400 fs could be obtained for InAlAs grown at a temperature as low as 150 °C.

There are striking similarities between this material and the more well-studied "low-temperature" GaAs. Both materials are generally grown at low temperatures (in the 150–300 °C range) under arsenic overpressure and thereafter exhibit semi-insulating properties and fast response times.[3] Since it is likely that a device will be fabricated on top of such a layer, the crystalline quality of these layers is of great importance.

In this letter, we show how the crystalline quality of In$_{0.52}$Al$_{0.48}$As/InP layers is a dramatic function of the growth temperature. Transmission electron microscopy (TEM) in plane view and cross section as well as particle-induced x-ray emission (PIXE) experiments have been carried out to study the influence of the growth temperature on the structure and on the amount of excess arsenic that can be incorporated in these layers.

A series of 1-μm-thick In$_{0.52}$Al$_{0.48}$As layers was grown on a Fe-doped semi-insulating (100) InP substrate by molecular beam epitaxy (MBE) at a growth rate of 0.7–1 μm/h and under As overpressure [As/(Al + In) flux ratio ~20]. The substrate temperature was then varied from one sample to the other from 150 to 450 °C. Growth was done on an As stabilized (2×4) reconstructed surface. One sample grown at 150 °C was further annealed *in situ* under As overpressure at 500 °C for 10 min after the growth was completed. The morphology of the samples was characterized by increased roughness at lower growth temperature. High resistivity was observed in all the layers. For the material grown at 150 °C, the resistivity further increased by approximately an order of magnitude after annealing.

The stoichiometry of the layers was measured by PIXE using a 0.45-MeV H$^+$ beam with the sample tilted at 70° with respect to the ion-beam direction. These parameters were chosen so that the P$_{K\alpha}$ signal, characteristic of the substrate, is not observable in the PIXE spectrum. Figure 1 is a representation of the results after they have been "normalized" to a reference sample grown at 450 °C. The variation of As content in the layers is evidenced by plotting the variations of the normalized [As]/[In], $k(T_g)$, for samples grown at different growth temperatures T_g. An obvious remark is that the relation between growth temperature and the incorporation of excess As in the layer is not simple. The same experiments on LT-GaAs clearly show a simple, monotonic relation between excess As incorporation in the layers and growth temperature.[4–6] Here, the general tendency is the same; i.e., for $T_g = 300$ °C, the $k(T_g)$ ratio is 1.07 and for $T_g = 150$ °C, $k(T_g) = 1.113$. The sample grown at 150 °C and then annealed *in situ* has lost some of its excess As but remains As-rich, with $k = 1.08$. The sample grown at 200 °C however, has a very low extra As content, with $k = 1.038$, and is clearly seen as an anomaly in the curve shown in Fig. 1.

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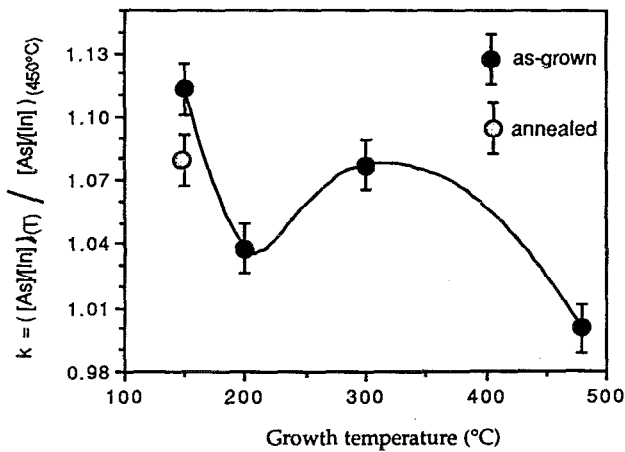FIG. 1. Arsenic concentration as a function of growth temperature obtained from PIXE experiments. The curve is an extrapolation to guide eyes.

FIG. 2. Set of cross-section TEM images (220 bright field images) showing the structure of the layers grown at 450 and 150 °C.

Cross-sectional samples for TEM were prepared by mechanical thinning to 30 μm, followed by ion milling. Plan-view samples were also prepared by chemical etching and occasional ion milling from the back side. TEM experiments were carried out on a JEOL 200 keV top-entry microscope with a resolution of 0.24 nm for imaging along [011] directions and on the Atomic Resolution Microscope (ARM) at Lawrence Berkeley Laboratory operating at 800 keV with a resolution of 0.17 nm for high-resolution imaging along $\langle 100 \rangle$ direction.

Figure 2 is a set of micrographs showing the dramatic change in the structure of the layers grown at different temperatures. The layer grown at 450 °C is a monocrystalline layer perfectly lattice matched on the InP substrate with no dislocation at the interface. However, stacking faults are sometimes found starting at some depth from the interface and extending toward the surface while forming microtwins. This fault is observed in all the layers grown down to $T_g = 200$ °C, indicating the relaxation of some residual stress during growth. For $T_g = 150$ °C, however, the structure of the layer is no longer monocrystalline, and pyramidal defects are clearly seen in high density. These pyramidal defects consist mostly of twin systems associated with dislocations. Electron diffraction and dark field experiments on plan-view samples show that, from the surface, these layers are seen as a mixture of InAlAs grains growing along the $\langle 100 \rangle$ and $\langle 011 \rangle$ directions. The average size of these grains is about 50 nm. Plan-view images also show that each grain is heavily faulted on its {111} planes. Additional spots in the diffraction pattern are readily identified as caused by rhombohedral arsenic (often referred to as hexagonal As for crystallographical convenience) with a particular orientation relationship with the InAlAs grains (Fig. 3). This orientation relationship is usually the same as that found for hexagonal As precipitates in GaAs.[7] High resolution electron microscopy (HREM) clearly shows the presence of a hexagonal structure compatible with As between adjacent InAlAs grains.[8] These regions may account for the very large amount of
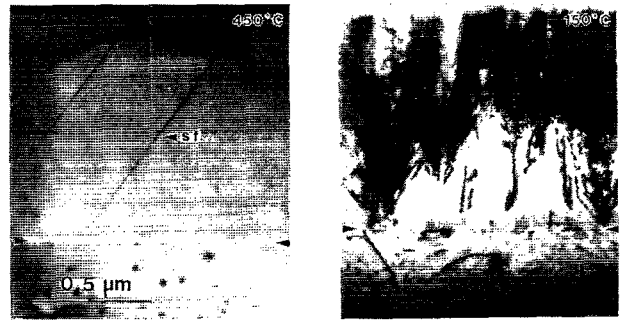
excess As measured by PIXE in these layers. These pyramidal defects have the same morphology as those observed in LT-GaAs.[5,9,10] As already discussed for LT-GaAs,[5,6,9] we think that this morphology, including the presence of hexagonal As found in the as-grown samples, is formed during growth. Because of the high concentration of excess As, the layer is heavily stressed. The usual stress-relieving mechanism, creation of dislocations on the surface that then move down into the layer by glide mechanisms, cannot operate in this case because of the low growth temperature. Roughening of the surface and As segregation are likely to occur on these surfaces. If we assume a mechanism similar to that proposed for LT-GaAs,[4,5] i.e., different growth rates for different facets and preferential ordering of basal hexagonal As planes along the {111} planes in InAlAs, the general structure of these defects may be explained.[8] However, we have clearly shown that the defects are due to the excess As in the layer. The lower the
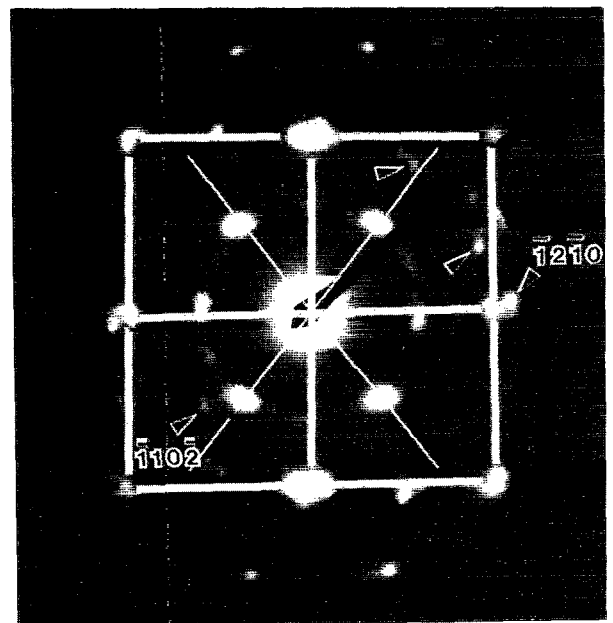

FIG. 3. Typical electron diffraction pattern obtained in plan-view layer along $\langle 100 \rangle$ substrate orientation. Arrows point toward spots due to hexagonal As.
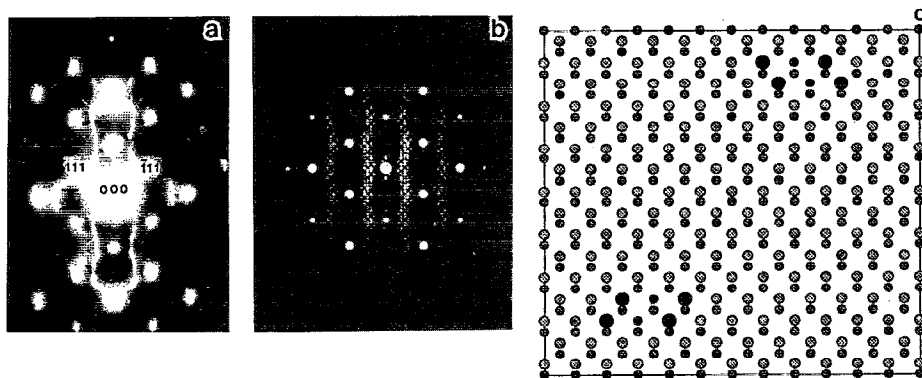
FIG. 4. (a) Experimental transmission electron diffraction (TED) pattern along $\langle 011 \rangle$ direction; (b) simulated TED pattern; (c) small ordered domains used for simulations.

growth temperature, the higher the amount of excess arsenic that can be incorporated. If this amount is too high, the increase of stress in the layer leads to the breakdown of monocrystalline growth.

The anomaly at $T_g = 200\,°C$ in Fig. 1 was also investigated by TEM. Figure 4(a) is a typical electron diffraction pattern obtained along [011] on the sample grown at 200 °C. Wavy diffuse scattering rods are seen running in the [100] direction. Intensity maxima are found at positions such as $h \pm 0.5$, $k \pm 0.5$, and $l \pm 0.5$. Such diffraction patterns may be explained by ordering the InAlAs alloy along two sets of {111} planes so as that the stacking sequence is As/In/As/Al... This structure is referred to as the Cu-Pt type on group III atoms and has already been observed in a number of ternary compounds.[11,12] To our knowledge, this is the first time that this ordering has been observed in a material grown in such far-from-equilibrium conditions. However, to create such wavy streaks in the diffraction pattern, ordered domains have to be small and of a special shape. We have performed extensive computer simulations[8,13] to deduce the average size of the domains. Figure 4(c) represents the "object" that was used to produce the simulation in Fig. 4(b). It is obvious that these ordered domains are very small since they contain about 20–30 properly ordered atoms. Note that in our simulations, it was not necessary to introduce antiphase domains[14] to get the wavy appearance seen in the diffraction pattern. The small amount of excess As incorporated in this layer as measured by PIXE, despite the low growth temperature of 200 °C, must be related to the presence of ordered domains in the layer. Ordered domains are usually formed on the surface during growth, then incorporated in the layer when the growth proceeds. Ordering the crystal by stacking separately the three different atomic constituents along two sets of {111} planes is thought to limit the incorporation of excess As in the layer.

In conclusion, our study shows that 1-μm-thick $In_{0.52}Al_{0.48}As$ layers can be grown on InP with a high crystalline quality for $T_g \geqslant 200\,°C$. The amount of excess As incorporated in the layers tends to increase when T_g is lowered, except for $T_g = 200\,°C$, where ordering along {111} planes occurs. Ordering of the material in small domains seems to prevent the layer from incorporating large amounts of As. At a lower growth temperature ($T_g = 150\,°C$), pyramidal defects are formed that mostly consist of twins, dislocations, and hexagonal As. It is thought that these defects are the result of stress induced

by As oversaturation. It is suspected that surface roughening may play a major role in the breakdown of monocrystalline growth in these layers. This study also clearly shows that the fast response that was previously evidenced in the layers grown at 150 °C (Ref. 2) must be related to either the presence of hexagonal As in the material, or more reasonably to the large density of defects such as dislocations that are observed in these layers.

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