Alloy inhomogeneities in InAlAs strained layers grown by molecular-beam epitaxy

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Transmission electron microscopy studies have been performed to characterize $In_xAl_{1-x}As$ layers grown by molecular-beam epitaxy on (100) InP substrates. The first observations of compositional nonuniformities in strained InAlAs layers are reported. The coarse quasiperiodic structure present in each sample has been found to be dependent upon the growth parameters and the sample characteristics.

Solid immiscibility is a widespread problem in ternary and quaternary III-V alloys. All III-V ternary alloys with lattice parameter (a) different to their binary end components ($\Delta a > 0$) have positive enthalpies of mixing, with the deviation from the ideality increasing with Δa^2 for disordered alloys.[1] Molecular-beam epitaxy (MBE) and organometallic vapor phase epitaxy (OMVPE) techniques can be used to grow these metastable alloys inside the region of solid immiscibility as a result of the kinetic limitations on the speed with which the constituents can redistribute themselves on the surface during growth. On this basis, the growth of III-V alloys over a complete range of compositions should be possible. However, it is well known that the strain state of the solid on which deposition is made can affect the growth product, even when the growth is homogeneous. If a compositionally modulated layer starts growing, the subsequent growth occurs not on the original unstrained substrate but on an inhomogenously strained material. Therefore, it is necessary to study this inhomogeneous layer to obtain information on the surface kinetic processes included in these growth techniques. In particular, it is necessary to know the ranges of temperature and composition in which modulation can occur. To our knowledge, although much work has been devoted to the study of these inhomogeneities in lattice matched or slightly mismatched layers of InGaAs,[2,3] InGaAsP,[4] and InAlAs[5] grown by different techniques, no proper contrast modulations have been observed in higher mismatched layers.[6]

In this work we report observations of compositional nonuniformities in strained $In_xAl_{1-x}As$ layers grown by MBE on InP (100) substrates. The composition and growth temperature of layers analyzed are shown in Table I. For electron microscopy, planar view specimens were prepared in the standard way by iodine ion milling. TEM observations were carried out at 200 kV with a HITACHI 800 NA microscope.

Figures 1, 2, and 3 show the microstructural features observed in $In_xAl_{1-x}As$ layers of different compositions ($x = 51.5$, 55.0, and 58.9 are shown in Figs. 1, 2, and 3, respectively). A coarse tweedlike structure was visible along the [001] and [010] directions using a ⟨220⟩ reflection. When viewed in ⟨400⟩ reflections, only the contrast band perpendicular to the g vector were visible, indicating strains present along the ⟨100⟩ directions. On average, the dark bands appear at a fixed separation, namely the wavelength of the quasiperiodic structure, Λ, the values of which are reported in Table I. The structure was found to have more accentuated boundaries in the compressive samples, B and C, than in the tensile sample A. There are also stacking faults observed in these samples.

We have shown previously[2,7] that increases in the potential energy of a mismatched epilayer-substrate material system as growth proceeds may be accommodated by the formation of modulations of composition of the layer. On the initiation of heteroepitaxial layer growth, strain is accommodated elastically and registry is maintained between the lattices of the epilayer and the substrate. As growth proceeds, further increases in the potential energy of the system may be accommodated by modulations in the composition of the layer. As the layer thickness increases, there is a decrease of the wavelength Λ of the modulation to absorb the corresponding increase in energy. When the layer thickness exceeds a certain value, the accommodation of energy by means of defect nucleation is energetically favorable to a further decrease in wavelength and defects start to propagate. In this last stage of the growth, relaxation occurs entirely by defect formation and the inhomogeneities are no longer apparent.[7]

To know the critical value at which defects begin to appear, an assessment of the elastic energy in the system including the energy associated with the nucleation of defects and composition modulation is necessary. In the case of slightly mismatched layers of InGaAs [misfit parameter ($f \approx 0.08\%$)], we have found a critical value of 0.5 μm.[8] By analogy with the case of homogeneous layers, we will call this value apparent critical layer thickness, t_{ca}. Beyond

TABLE I. Growth temperatures, layer thicknesses, and wavelengths of the coarse pattern for the samples studied.

	x	T	Λ (nm)	Thickness (μm)
A	51.5	580 °C	600	1
B	55.0	580 °C	460	1
C	58.9	570 °C	820	2

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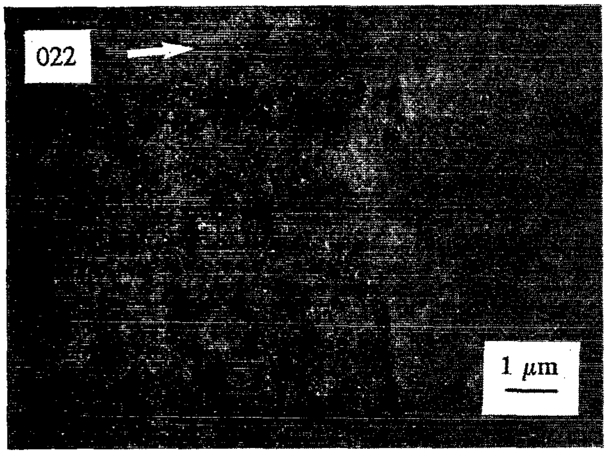

FIG. 1. Plan view $g = 022$ micrograph of sample A. The contrast bands are slightly visible.

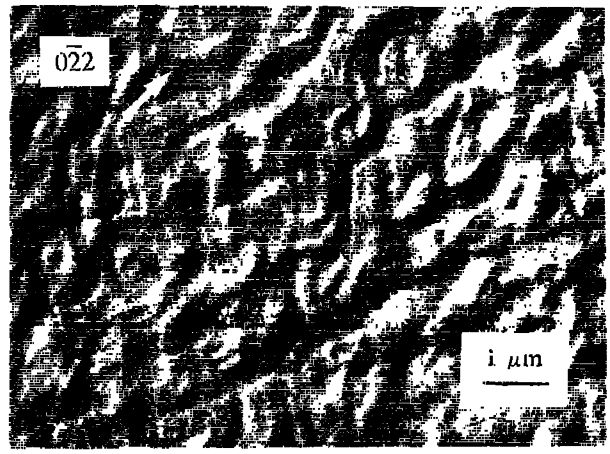

FIG. 3. Plan view micrograph of sample C under $g = 0\overline{22}$. The dark bands are in stronger contrast in compressive samples.

this value, the nucleation and propagation of defects, such as stacking faults and threading dislocations, is favored, rather than a further reduction of the coarse period.

In this work, taking into account that all samples analyzed have thicknesses greater than 1 μm and misfit parameter f is between 0.05% and 0.5%, we assume the layer thickness to be beyond the t_{ca} value. Under these conditions, the presence of stacking faults may be explained by the large thicknesses of the samples studied. We observe that for samples of identical thickness (A and B) the wavelength is smaller for the more mismatched sample. We suggest that the observation of the quasiperiodic structure depends not only on the growth conditions (temperature) but also on the strain (thickness and mismatch). This structure appears at the initial stages of growth but its evolution is dependent on the growth parameters and sample characteristics. On a macroscopic level it is known that the substrate influences the composition of an homogeneous overgrowth by favoring layers with a lattice parameter equal to its own. This is the "pulling effect."[9] According to Glas[10] this effect also exists at the microscopic level:

FIG. 2. Coarse structure and stacking faults in sample B, imaged with $g = 022$.

the zones of the surface of the layer having a large (respectively, smaller) lattice parameter will favor the growth over them with a larger (respectively, smaller) lattice parameter. For a given atom, because of the modulated deformation of the surface, the difference of the potential $\Delta\mu$ between the gas phase (in MBE) and the surface (which is the driving force for the growth), will be modulated along this surface. Thus, once a composition modulation has started at some stage of the growth, it may continue. At this point our results disagree with the conclusions of Glas who said that the modulation will continue with the same period and that only periods of about 100–200 nm tend to develop. What remains unexplained is how such a modulation starts, though it may be due to energetic or kinetic factors, such as strain induced in the substrate by nuclei at the onset of growth, the nucleation rate, or surface diffusion.

In summary, our results show that the coarse structure associated with compositional modulation are also present in $In_xAl_{1-x}As$ strained layers grown by MBE on InP substrates. The wavelength of these structures is dependent upon the growth conditions, alloy composition, and thickness of the layer.

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