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# Indium diffusion in the chemical potential gradient at an $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{In}_{0.52}\text{Al}_{0.48}\text{As}$ interface

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We have measured the distribution of group III metals at  $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{In}_{0.52}\text{Al}_{0.48}\text{As}$  interfaces before and after annealing at 1085 K. We find little evidence for Al interdiffusion, but the Ga concentration profiles show some broadening on annealing. Also, the originally nearly constant In profiles develop strong modulations with near discontinuities at the original interfaces. This phenomenon is explained and modeled in terms of In diffusion in the chemical potential gradient established by the disparity of the Al and Ga mobilities and the requirement of III-V stoichiometry in the alloys.

We have measured, by Auger compositional depth profiling, the interdiffusion of group III metals at annealed  $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{In}_{0.52}\text{Al}_{0.48}\text{As}$  interfaces. We find, rather than the simple Al/Ga interdiffusion that had been assumed in a prior study,<sup>1</sup> a more complex behavior in which In and Ga interdiffuse against an essentially constant Al concentration profile. This behavior, which leads to a near discontinuity in the In concentration at the location of the original interface, can be rationalized if one assumes that the mobilities of the group III metals are inversely related to their bonding strength in the lattice, i.e., the mobilities are ordered  $\text{In} > \text{Ga} > \text{Al}$ . In this case, Ga diffusion is driven by the Ga concentration gradient; and In, which is more mobile than the Al, diffuses in the chemical potential gradient established by the disparity between the Ga and Al mobilities and the requirement of stoichiometry. This effect is general to interdiffusion at multicomponent interfaces where the mobilities of the diffusing species differ. A similar effect was first observed in solids by Darken for the interdiffusion of carbon and silicon in iron.<sup>2</sup>

The interdiffusion experiments were carried out on  $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{In}_{0.52}\text{Al}_{0.48}\text{As}$  heterostructures grown by molecular beam epitaxy (MBE) on semi-insulating, Fe-doped InP substrates. The structures were similar to those used by Chang and Koma in a study of the interdiffusion of Ga and Al at the GaAs/AlAs interface.<sup>3</sup> They consisted of a single 100 nm layer of  $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$  in an  $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$  matrix. The samples were uniformly lightly doped with Si to provide electrical conductivity which prevented sample charging during Auger depth profiling. Isothermal annealing of the samples was carried out in a horizontal furnace under flowing nitrogen at 1085 K. Loss of the volatile group V elements during high-temperature long-term annealing was prevented by using a "pill-box" arrangement to provide group V overpressure. The "pill box" was made using a quartz ring (1 mm thick, 8 mm diameter) with both ends polished. The ring was enclosed at both ends with GaAs wafers, and the sample was placed in the resulting cavity.

The gap between the sample surface and the top GaAs wafer was about 0.5 mm. The Auger compositional depth profiles were carried out in a Physical Electronics 545 scanning Auger microprobe that has been modified by the addition of a vacuum interlock sample introduction system and a computer-based data acquisition system. The Auger spectra were excited with a 3 kV primary electron beam and detected in derivative mode with 4 V modulation on the analyzer. The depth profiles were produced by ion sputtering with a rastered 2 kV argon ion beam. We estimate the depth resolution of this experiment in the vicinity of the interfaces to be on the order of 10.0 nm (10%–90%).<sup>3,4</sup> This uncertainty in depth resolution is on the same order of magnitude as the diffusion lengths in these experiments, and constitutes the major source of error. Annealing for longer times or at higher temperatures, under these experimental conditions, leads to surface roughening which further degrades depth resolution.

Figure 1 shows the compositional depth profiles derived from Auger peak-to-peak height versus sputter-time data. The known thicknesses of the MBE-grown layers provided an internal reference for the conversion of sputter time to distance. Figure 1(a) shows composition versus distance from the surface for an as-grown sample. The Ga and Al concentrations exhibit abrupt changes at the interface, and the apparent width of the interface is limited by the resolution of the sputter profiling experiment. The In concentration is essentially constant except for small negative excursions just outside the interfaces. These features are the result of flux transients produced by opening the Ga or Al shutters during the MBE growth procedure. On opening a shutter, a brief period of high Ga or Al flux is produced which causes the growing layer to be momentarily rich in Ga or Al and deficient in In. The spike of Ga or Al at the interface, which lies on the edge of an abrupt concentration change, is obscured by the resolution of the experiment, while the dip in the constant In concentration merely broadens and remains visible.

Figure 1(b) shows the concentration profile measured after annealing a structure identical to that in Fig. 1(a) for 10 h at 1085 K. The Al profile is essentially unchanged, but

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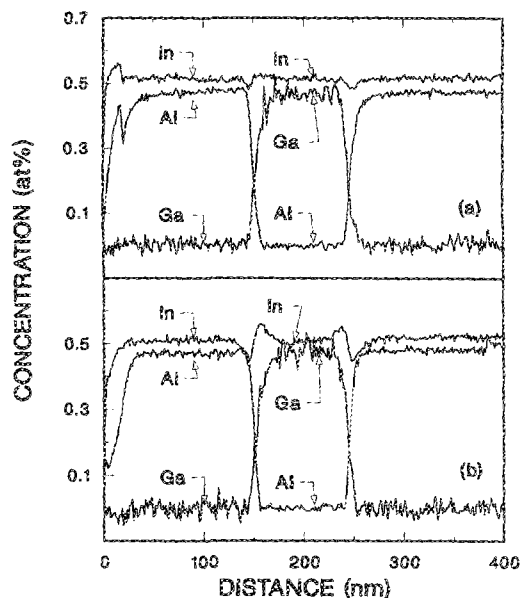


FIG. 1. Auger compositional depth profiles showing the distribution of group III elements at MBE-grown  $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{In}_{0.52}\text{Al}_{0.48}\text{As}$  interfaces. The Auger peaks measured were  $\text{Al}_{\text{LMM}}$  at 68 eV,  $\text{In}_{\text{MNN}}$  at 404 eV, and  $\text{Ga}_{\text{LMM}}$  at 1070 eV. (a) as grown; (b) annealed for 10 h at 1085 K.

the Ga profile has spread out relatively to the Al profile. More dramatically, the In profile has developed a strong symmetric modulation with abrupt changes at the interfaces. The width of the Ga interfaces and the width of the In modulations are comparable.

The interfacial concentration profiles in Fig. 1(b) can be approximated in terms of a simple model. If we assume that the mobility of the Al is zero, and that the III-V stoichiometry must be maintained, then the system can be characterized by a single effective diffusion coefficient  $D_{\text{eff}}$ , which describes the mobility of the Ga and the In. In the time regime when the diffusion lengths are less than half the hetero-layer thickness, we can neglect the presence of the other interface. Then, letting  $x$  be the distance from the original interface, and  $C_{\text{Al}}$ ,  $C_{\text{Ga}}$ , and  $C_{\text{In}}$  be the concentrations of Al, Ga, and In, respectively, the concentrations as a function of time,  $t$ , and distance can be written for the left interface as

$$C_{\text{Al}} = 0.48 \quad \text{for } x < 0.0 \quad (1)$$

$$= 0.00 \quad \text{for } x > 0.0,$$

$$C_{\text{Ga}} = 1/2 (0.47) [1.0 - \text{erf}(x/\sqrt{4D_{\text{eff}}t})], \quad (2)$$

$$C_{\text{In}} = 1.0 - (C_{\text{Ga}} + C_{\text{Al}}). \quad (3)$$

Equation (1) states that the Al profile does not change with time, Eq. (2) is the well known result for diffusion from an initially abrupt interface into an infinite solid,<sup>5</sup> and Eq. (3) is the stoichiometry boundary condition. Figure 2(a) shows the calculated concentration profiles plotted as a function of the reduced distance ( $x/\sqrt{4D_{\text{eff}}t}$ ). This model contains the essential features that the Al concentration remains sharp, the Ga profile spreads out, and the In concentration becomes modulated. When the diffusion length becomes comparable to half the layer thickness, the well known results for diffu-

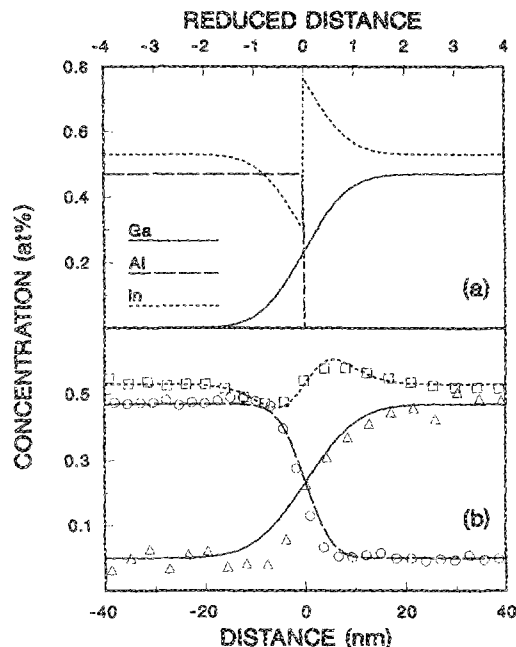


FIG. 2. (a) Calculated group III metal distributions at an annealed  $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{In}_{0.52}\text{Al}_{0.48}\text{As}$  interface for  $D_{\text{eff}} = 7 \times 10^{-18} \text{ cm}^2/\text{s}$  and  $t = 36000 \text{ s}$ . The scale at the top of the figure is in reduced distance, ( $x/\sqrt{4D_{\text{eff}}t}$ ). (b) Profiles from (a), broadened by convolution with a Gaussian of 10 nm FWHM to simulate the experimental depth resolution, plotted over the experimental data points [ $\square$ ] = In,  $\circ$ ] = Al, and  $\triangle$ ] = Ga]. Only every other data point is shown for clarity.

sion from an extended source of limited extent can be used to describe the profiles.<sup>5</sup>

In Fig. 2(b) the concentration profiles, convoluted with a Gaussian of 10.0 nm full width at half-maximum to simulate experimental broadening, are plotted versus distance assuming a  $D_{\text{eff}} = 7 \times 10^{-18} \text{ cm}^2/\text{s}$  and  $t = 36000 \text{ s}$ . These profiles essentially reproduce the measured profiles which are plotted as points. This value of  $D_{\text{eff}}$  is comparable but somewhat larger than both the self-diffusion coefficient of Ga in GaAs ( $D_{1085} = 1 \times 10^{-19} \text{ cm}^2/\text{s}$ )<sup>6</sup> and the interdiffusion coefficient of Ga and Al at the GaAs/AlAs interface ( $D_{1085} = 3 \times 10^{-19} \text{ cm}^2/\text{s}$ ).<sup>3</sup> The model can be generalized at the expense of the clear physical picture that Eqs. (1)–(3) convey.<sup>7</sup> However, since the lattice constant of this alloy system depends on the In concentration, the change in volume and associated layer strain will complicate a complete description of the chemical potential of this system.

It should be noted that the effect described in this letter requires a difference in mobility between those elements which exhibit a discontinuity at the interface (Ga and Al in this case), and that this phenomenon has been observed for annealing times which are long relative to those that have been used in other experiments.<sup>1</sup> For very short time anneals it may be possible for transient effects, such as an accumulation of dislocations or other lattice defects at the as grown interface, to equalize the mobilities of the Ga and Al and thus to minimize the establishment of a chemical potential gradient for the In. The limitations of present depth profiling techniques make it difficult to directly observe the concentration profiles in the short time anneal regime. However, the possibility of this "Darken-like" diffusion should be con-

sidered when modeling the electrical or elastic properties of annealed multicomponent interfaces.

We would like to thank P. R. Berger for growing the MBE heterostructures used in these experiments and to acknowledge helpful discussions with K. S. Seo and J. Singh and with R. C. McCune who pointed out the similarity of these observations to the "Darken experiment."<sup>2</sup> Also, two of us GPK and PKB would like to acknowledge the support of the Department of Energy under grant DE-FGO2-86ER45250.

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