Effect of alloy clustering on the high-temperature electron mobility in $In_{1-x}Ga_xAs_yP_{1-y}$

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Hall measurements on liquid phase epitaxial $In_{1-x}Ga_xAs_yP_{1-y}$ lattice matched to InP have been performed in the temperature range $300 < T(K) < 600$ °C. The crystals were grown at 640 °C. Anomalous lowering of the mobility and carrier concentration has been observed at these temperatures for certain alloy compositions. It is believed that clustering due to miscibility gaps existing in these solid compositions is responsible for the observed data. Interpretations of the data based on this assumption have been made.

Alloy clustering in III-V ternary and quaternary compound semiconductors results from the random or near-random distribution of mixed elements in cation or anion sublattice sites. In general, its effect on the material properties is taken into account by an additional carrier-scattering mechanism in the crystal. The effects of alloy clustering are very much dependent on the size of the cluster and effects dependent on the latter have been observed in the compounds and in devices made from them.[1–6] Clustering and associated composition modulations have been observed in certain composition ranges of $In_{1-x}Ga_xAs_yP_{1-y}$ lattice matched to InP.[7,8] The modulations are of the order of 100 Å and the corresponding composition deviations are about ± 10%. For liquid phase epitaxial growth at temperature of about 650 °C, a miscibility gap has been predicted for a certain composition range of the quaternary alloys,[9,10] leading to instability and spinodal decomposition in the solid phase. Growth of single layer crystals is possible, however, due to strain induced by the substrate. The strain energy in these alloys has been calculated by de Cremoux[11] and Srinivasa and Bhattacharya.[12] Clustering, which is a consequence of the miscibility gap, leads to lower mobilities in the crystals and it has been demonstrated that a 20–25% mobility enhancement can be obtained by growing the layers at elevated temperatures.[4] In this communication we will present anomalous Hall mobility data on $In_{1-x}Ga_xAs_yP_{1-y}$ recorded for lattice temperatures greater than 300 K. The data have been analyzed and it is shown that the behavior arises from clustering and associated effects.

Undoped layers of the quaternary alloys, ranging in composition from InP to $In_{0.53}Ga_{0.47}As$ were grown on (100)-oriented InP substrates by liquid phase epitaxy. Growth was accomplished in a horizontal graphite slider boat. The data reported here are for layers grown at 640 °C using the step-cooling technique. The mixed crystal compositions were derived from photoluminescence measurements. The lattice mismatch in the layers was within ± 0.03%. Typical layer thicknesses were 4–8 μm.

Hall measurements were made on symmetrical samples using the van der Pauw method over a temperature range of 20–500 K. This communication is concerned with data for

$T > 300$ K. The variation of Hall mobility with temperature for samples with varying composition is shown in Fig. 1. The data have been analyzed by taking into account ionized impurity, polar optical phonon, alloy, deformation potential, piezoelectric, and space-charge scattering mechanisms. The solid lines close to data points for each composition represent the theoretically calculated mobilities. Several anomalous features may be mentioned. The value of the alloy scattering potential ΔU needed for the analysis is unusually high and ranges between 0.6–0.8 eV for crystals with compositions $0.5 < y < 0.9$ ($y \simeq 2.16x$ for lattice-matched compositions). Similar values have been calculated and reported by other authors,[13] but such values are much larger than values of ΔU estimated from electron affinity or electronegativity differences.

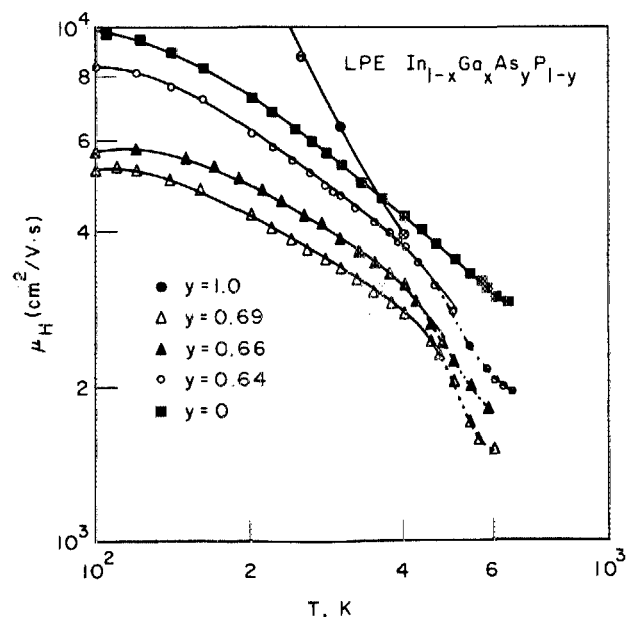The second and more interesting feature observed in the

FIG. 1. Variation of Hall electron mobility in $In_{1-x}Ga_x/As_yP_{1-y}$ with temperature (with $y \simeq 2.16x$). The solid curves indicate mobility values computed from the relevant scattering mechanisms. The significant lowering of the mobilities with increase of temperature, which is in variation with theoretically calculated data, is evident for some alloy compositions.
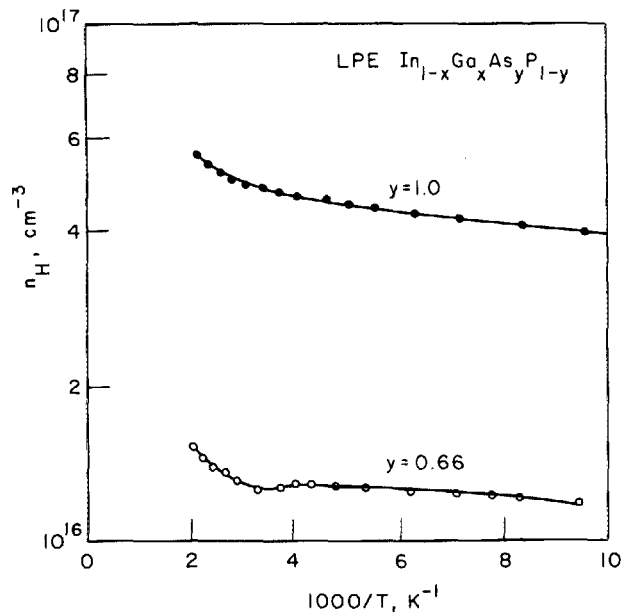
FIG. 2. Variation of Hall electron concentration with temperature for two samples with $y = 0.66$ and 1.0 ($x = 0.31$ and 0.47, respectively).

data is an abrupt lowering of mobility for $T > 370$ K in crystals with $y \sim 0.65$. This behavior, which is not observed in crystals of compositions near InP or $In_{0.53}Ga_{0.47}As$, cannot be explained by theoretical analysis. In other words, no reasonable combination of parameters for the different scattering mechanisms can explain the observed trend in mobility. An associated effect observed in the temperature dependence of Hall electron concentration n_H for the same samples is shown in Fig. 2. The noticeable dip observed in these samples was not observed for other compositions. The possibility of the substrate contributing to these effects was checked by thinning the substrates in these samples and repeating the measurements. The same behavior was observed. The substrate-layer interface may be playing a role, but it is believed that the observed effects reflect, at least in part, properties of the grown epitaxial layer.

Since the compositions for which the anomalous results are observed coincide with those for which a miscibility gap and associated instability are predicted, it may be assumed that clustering is in some way responsible. One then has to describe a plausible mechanism which can explain the data. Blood and Grassie[14] have recently postulated a model in which the boundary of a cluster region is assumed to be analogous to a heterojunction discontinuity. By virtue of the relative extents of the bandbending and the clusters, the conduction band edge shows a periodic modulation with peaks and troughs separated by ΔE_c, the conduction band discontinuity. These authors have calculated the value of ΔE_c to be ~ 0.08 eV. So for $T > 400$ K there can be an additional component of scattering due to the interaction of electrons with the potential distribution forming the peak. If this scattering

mechanism has a temperature-dependent mobility $\mu \propto T^{-n}$, where n increases with T, the observed mobility data at high temperatures can be explained. The anomaly in the n_H vs T data cannot be explained with this model.

The anomalous $\mu_H(T)$ and $n_H(T)$ behavior can be explained by considering thermal transfer of electrons from the conduction minimum to higher lying states, in a manner somewhat similar to intervalley transfer of carriers. In fact, large clusters of localized defects can cause asymmetric fluctuations of the impurity as well as band-edge potential and give rise to conduction states of higher density above the minimum. Using the analysis of Blood,[15] and Aukerman and Willardson[16] with the $n_H(T)$ data, a value of 0.091 eV is obtained for the energy separation of such states from the conduction minimum. The sharp fall in the mobility with increasing temperature can then be thought to be due to a carrier transfer effect.

Finally, another alternate explanation for the observed behavior of the transport parameters at high temperatures can be offered by invoking the effect of clusters on optical-phonon modes in the crystal. Clusters cause the crystal potential to fluctuate throughout the crystal. Calculations by Verleur and Barker[17] indicate that LO phonon frequencies may be lowered with clustering. This could cause carrier-mobility lowering at higher temperatures, as observed in our measured data.

In conclusion, effects in the high-temperature Hall data, which probably arise from the existence of a miscibility gap and associated clustering, have been presented. The data reflect carrier transfer and scattering effects caused by conduction-edge potential modulations.

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