Nonrandom alloying in $In_{0.52}Al_{0.48}As$/InP grown by molecular beam epitaxy

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Anomalous behavior in Hall data of molecular beam epitaxial $In_{0.52}Al_{0.48}As$/InP for $T \geqslant 400$ K suggests the presence of alloy clustering. The relaxation time for alloy clustering has been determined and analysis of measured data gives an estimate of composition fluctuation and cluster size. Evidence of clustering in the same samples is also obtained from the measured velocity-field characteristics, low-temperature photoluminescence data, and carrier impact ionization coefficients.

Due to recent interest in the $In_{0.52}Al_{0.48}As$/$In_{0.53}Ga_{0.47}As$ heterostructure system for high-speed optoelectronic and electronic devices, the quality of InAlAs grown by molecular beam epitaxy (MBE) is of importance. Experiments based on photoluminescence and carrier transport indicate that the system is not of a high quality and may have alloy clustering present.[1,2] Although a periodic short-range order as that present in superlattices can be advantageous, clustering generally has a detrimental effect on the optical and transport properties of materials.[3–5] Due to the large difference between In- and Al-related bond energies the $In_{0.52}Al_{0.48}$ As system is expected to show clustering due to the low temperatures employed in MBE growth. In this study we have made temperature-dependent Hall measurements on Si-doped $In_{0.52}Al_{0.48}$ As grown at 500 °C and we find that the mobilities increase with increase of measurement temperature in the range of 400–600 K. To explain this behavior, we have developed a theoretical formalism for the scattering rate due to alloy clustering. In particular, this scattering can be dominant when the cluster size is much larger than the radius of the Wigner–Seitz cell (~ 2.5 Å).

Undoped and Si-doped $In_{0.52}Al_{0.48}$ As single layers were grown lattice matched to (001) InP:Fe in a three-chamber Riber 2300 epitaxy system. The mismatch in the layers was of $\sim 10^{-3}$–10^{-4}. A 0.4-μm undoped buffer layer was usually grown before growing the Si-doped layers. Typical growth rates and layer thicknesses were 1.3 μm/h and 1.5 μm, respectively. Hall measurements were made on photolithographically defined van der Pauw samples over the temperature range of 60–600 K. The room-temperature electron density in the samples studied varied in the range $(0.5–5.0) \times 10^{17}$ cm^{-3}. It was observed that for samples in which the mobilities were low, the mobility increased with the increase of temperature in the range of 400–600 K. This trend for two samples is depicted in Fig. 1.

The Hall data have been analyzed by taking into account ionized impurity, polar optical phonon, random alloy, deformation potential, and piezoelectric scattering mechanisms. The material parameters of $In_{0.52}Al_{0.48}$ As used in this calculation and taken from Ref. 6 are listed in Table I. Several anomalous features may be mentioned. The experimentally determined room-temperature mobilities of samples with carrier concentrations in the range of $(1–5) \times 10^{17}$ cm^{-3} are much less than calculated values even for a high compensa-

tion ratio. The second and more interesting feature observed in the data, as mentioned earlier, is that the mobilities increase with the increase of temperature for $T > 400$ K. This unusual behavior cannot simply be explained. Thus it is necessary to identify and study the scattering mechanism related to the alloy quality and dominant for $T > 200$ K.

Compositional inhomogeneities can lead to this behavior which has been observed by one of us earlier in the InGaAsP alloys, where also a miscibility gap is known to exist.[7] Marsh[3] has developed a formalism involving alloy clustering to explain velocity-field characteristics of $In_{0.53}Al_{0.47}As$. The scattering rate due to alloy fluctuations (alloy clustering) is given by matrix elements of the form

$$M(k,k') = \frac{1}{V_c} \int e^{i(k-k')r} V(r) dr, \qquad (1)$$

where

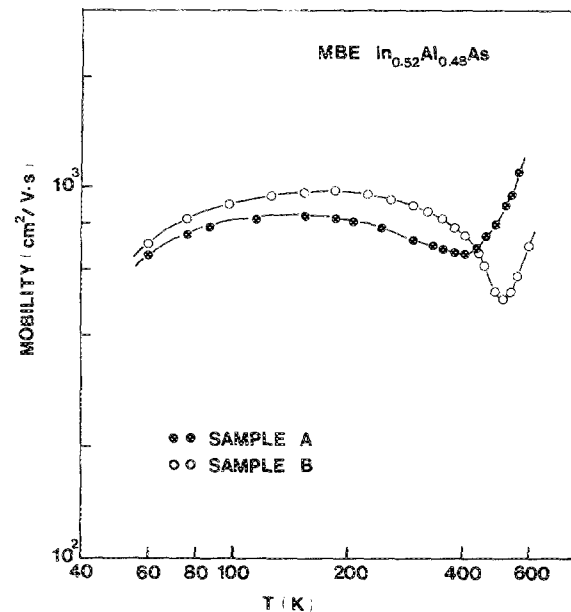$$\begin{aligned} V(r) &= V_0 & |r| \leqslant r_c \\ &= 0 & |r| > r_c \end{aligned}$$

FIG. 1. Temperature-dependent Hall mobility in Si-doped $In_{0.52}Al_{0.48}$As.

TABLE I. Materials constants for $In_{0.52}Al_{0.48}As$ used in the transport calculations.

Effective mass	$0.08m_0$
Optical phonon temperature	432 K ($= 37.4$ meV)
Static dielectric constant	12.7
Dynamic dielectric constant	10.3
Velocity of sound	4.4×10^5 cm/s
Mass density	4.7×10^3 kg/m^3
Acoustic deformation potential	5.8 eV
Lattice constant	5.8687 Å

and r_c is the radius of the cluster. $2V_0$ is the energy difference resulting from alloy fluctuations and V_c is the crystal volume. Thus scattering rate can be calculated by using Fermi's second golden rule:

$$\frac{1}{\tau(k)} = \frac{V_c}{(2\pi)^3} N_A \frac{2\pi}{\hbar} \int |M(k,k')|^2 \delta(E_k - E_{k'})dk'. \quad (2)$$

Substituting Eq. (1) into Eq. (2) and integrating gives the alloy scattering rate as

$$\frac{1}{\tau(k)} = \frac{4\pi m^*}{\hbar^3} (V_0^2 r_c^3) \frac{1}{2kr_c}$$

$$\times \left(1 - \frac{1}{(2kr_c)^2} + \frac{\sin(4kr_c)}{(2kr_c)^3} - \frac{\sin^2(2kr_c)}{(2kr_c)^4}\right). \quad (3)$$

The electron mobility corresponding to the scattering rate given by Eq. (3) is

$$\mu = \frac{e\langle\tau\rangle}{m^*} = \frac{e}{m^*} \frac{\langle k^2 \tau(k)\rangle f}{\langle k^2\rangle f} \quad (4)$$

and can be calculated numerically assuming a Maxwell–Boltzmann distribution function with the change of cluster size r_c. The potential height V_0 is kept constant.

The results for various cluster sizes are shown in Fig. 2. It can be seen that a cluster of up to 20 Å radius results in an approximate $T^{-1/2}$ dependence of μ_{Al}.[8] However, when the cluster size is larger than 25 Å ($kr_c \gg 1$), the high-temperature mobilities increase with the increase of temperature. With radii larger than 100 Å, mobilities start increasing from very low temperatures (~ 20–50 K). Taking into account the scattering due to alloy clustering, we have calculated the variation of mobility with temperature. The calculated mobilities are shown in Fig. 3 along with measured data. Since ionized impurity scattering and alloy scattering are dominant for $T < 100$ K and $T > 200$ K, respectively, the compensation ratio and the alloy fluctuation ($2V_0$) can be estimated from the fitting. Fairly good agreement with measured data in the temperature range 60–400 K is obtained by considering 35-Å clusters of $In_{1-x}Al_xAs$ with $x = 0.46$ to 0.50. The required alloy fluctuation ($2V_0$) is about 120 meV. The disagreement at higher temperatures is possibly due to ionizing carriers from a deep donor (~ 0.5 eV) in the alloys, producing a screening effect. Other than this we do not believe that the temperature-dependent mobility data are affected by deep levels. We have made systematic measurements on deep level defects in the ternary alloy,[9] and it was found that the deep level densities are at least two orders lower than the carrier concentrations in our samples. The
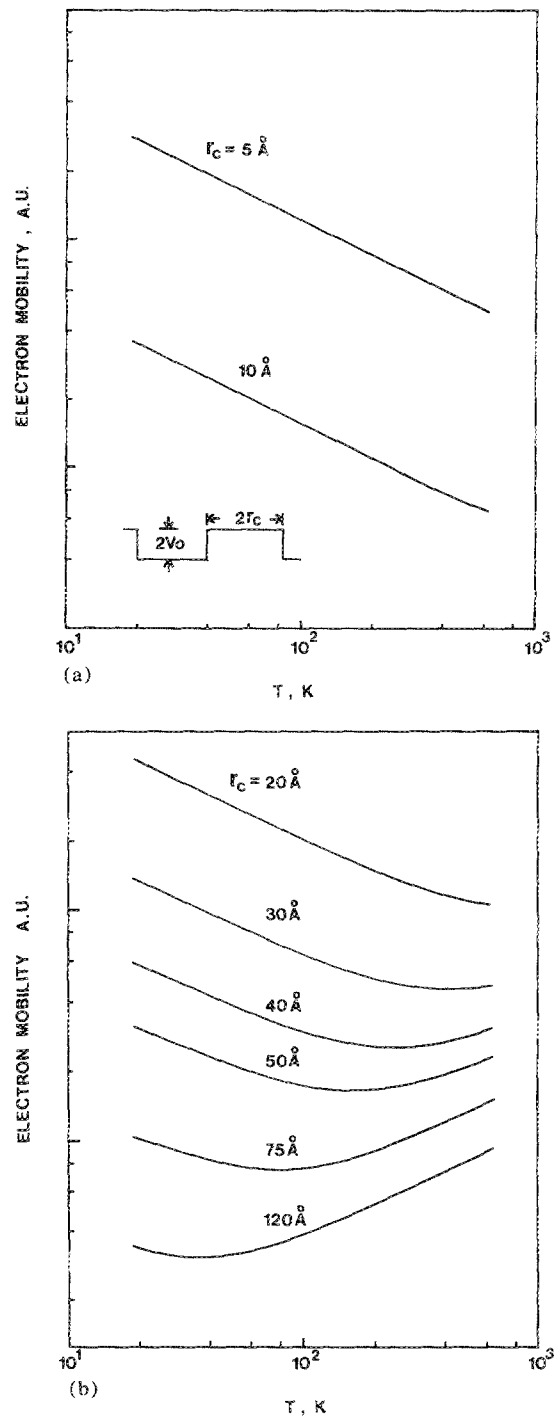

(a)

(b)

FIG. 2. Calculated temperature dependence of alloy scattering mobilities due to alloy clustering with the variation of cluster size (r_c) in $In_{0.52}Al_{0.48}As$ (V_0 is kept constant).

excitonic linewidth for a perfect $In_{0.52}Al_{0.48}As$ system (no clustering) is calculated to be ~ 4 meV and the linewidth increases with clustering, dependent on the growth conditions. The linewidth of the main peak in the 10-K photoluminescence spectra of one of our samples grown at 0.5 μm/h is ~ 19 meV. This suggests that the crystals either have a considerable amount of clustering or there might be a high density of native defects present in them. Alloy scattering described by Eq. (3) has been included in a Monte Carlo program to simulate the carrier velocity-field characteris-
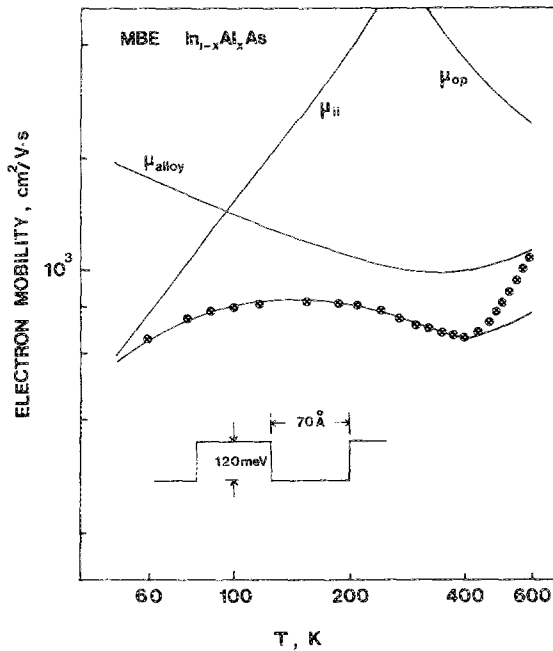
FIG. 3. Measured (●) and calculated (solid curve) temperature dependent Hall data on sample A. μ_{ii} and μ_{op} are the mobilities limited by ionized impurity scattering and optical phonon scattering, respectively. The inset shows the composition modulation resulting from alloy clustering and obtained from fitting of data.

tics.[10] The simulation shows that the peak velocities decrease with increase of clustering ($kr_c \gg 1$). We have also measured the velocity-field curves in the alloys by a pulsed current-voltage technique. The measured peak velocity v_p in a sam-

ple in which the 300-K mobility is 750 cm^2/V s is 6×10^6 cm/s. The calculated v_p in the absence of clustering is 1.4×10^7 cm/s. The cluster parameters used to explain low-field mobility data also explain this discrepancy. Measurement of the impact ionization coefficients in $In_{0.52}Al_{0.48}As$ by ensuring pure electron and hole injection gives $\alpha/\beta \sim 4$. This result, which agrees with the data of Capasso et al.,[11] can be explained by considering a modulation of the threshold ionization energies caused by a periodic potential variation ($2V_0$) due to compositional clustering.[12]

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