

GROWTH AND PROPERTIES OF $\text{In}_{0.52}\text{Al}_{0.48}\text{As}/\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$, GaAs:In AND InGaAs/GaAs MULTILAYERS

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We have investigated the properties of some In-containing materials and heterostructures grown by molecular beam epitaxy on GaAs and InP substrates. Photoluminescence spectra of InGaAs/InAlAs quantum wells have been related to growth kinetics and it is seen that growth interruption or the incorporation of a few periods of superlattices at the heterointerfaces smooths the growth front. Experiments have been performed to determine the properties of $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$ grown on InP. This lattice-matched alloy $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$ may be clustered under normal growth conditions at a substrate temperature $\sim 500^\circ\text{C}$. Addition of small amounts of In (0.2–1.2%) to GaAs reduces trap and defect densities in these materials, as seen from DLTS and low-temperature photoluminescence data. The improvement may be related to the higher surface migration rate of In compared to Ga and a subsequent reduction of point defects. Single-mode optical guides and direction couplers with losses as low as 1–3 dB/cm have been fabricated with GaAs:In and $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ strained-layer superlattices.

1. Introduction

$\text{In}_x\text{Ga}_{1-x}\text{As}$ and $\text{In}_x\text{Al}_{1-x}\text{As}$ are important alloy semiconductors for electronic and opto-electronic heterostructure devices. Together with $\text{In}_x\text{Ga}_{1-x}\text{As}-\text{GaAs}$ strained-layer superlattice (SLS), a large and important spectral energy range can be covered. Molecular beam epitaxy (MBE) is a growth technique which is principally controlled by surface kinetics, and therefore, the electrical and optical properties of the grown films and heterointerfaces can be non-ideal. The intent of this paper is to highlight some electrical and optical properties in these alloys and heterostructures, which can result from the growth kinetics. More specifically, results of photoluminescence measurements on $\text{In}_{0.52}\text{Al}_{0.48}\text{As}/\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ single quantum wells lattice matched to InP substrates suggest clustered growth in the Al-containing alloy and interface roughness at the heterointerface under normal MBE growth conditions. Results from high temperature Hall and carrier impact ionization measurements are presented, which also indicate the presence of clustering in the InAlAs alloys. Finally, the growth of $\text{In}_x\text{Ga}_{1-x}\text{As}$ ($x \leq 0.01$) and $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ SLS on GaAs substrates and the properties of optical components made with these materials are described.

2. Molecular beam epitaxial growth

The different materials and heterostructures were grown in a three-chamber RIBER 2300 epitaxy system. Extensive source and system baking were carried out [1] to grow high-purity materials. Undoped 120 Å GaAs/ $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ single quantum wells with 0.3 meV excitonic linewidths in the 2 K photoluminescence spectra and lattice-matched undoped $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ with $\mu_{300\text{K}} = 12,050 \text{ cm}^2/\text{V}\cdot\text{s}$ and $\mu_{77\text{K}} = 53,000 \text{ cm}^2/\text{V}\cdot\text{s}$ assert the purity of the growth ambient. In-doped GaAs and the InGaAs/GaAs SLS were grown on undoped or doped GaAs substrates at $560\text{--}580^\circ\text{C}$ at a growth rate of 1.2 $\mu\text{m}/\text{h}$. InAlAs and InGaAs/inAlAs were usually grown in the temperature range $480\text{--}520^\circ\text{C}$ on (001) Fe-doped semi-insulating or S-doped N^+ InP substrates at 0.5–1.3 $\mu\text{m}/\text{h}$.

3. Photoluminescence in $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{In}_{0.52}\text{Al}_{0.48}\text{As}$ single quantum wells

Photoluminescence measurements were done at 10 K and higher temperatures and with variable excitation intensity. The luminescence was excited with a 6328 Å He–Ne laser and was analyzed with

a 1 meter Jarrell–Ash spectrometer. Three categories of SQW samples were characterized. These are (i) samples grown with no interruption at the normal (*InAlAs* on *InGaAs*) and inverted (*InGaAs* on *InAlAs*) heterointerfaces, (ii) samples grown with up to 3 min growth interruption at the heterointerfaces, and (iii) incorporation of a three-period superlattice ($L_z = L_B = 15 \text{ \AA}$) at the heterointerfaces without interruption. The typical low-temperature photoluminescence spectrum is dominated by an intense peak at $\sim 0.81 \text{ eV}$, which is due to bound exciton transitions. As seen in fig. 1, the linewidth of this peak (FWHM) for a 120 Å SQW monotonically decreases with increasing periods of growth interruption at the heterointerfaces. This is due to a gradual smoothing of the growth front [2]. The linewidth varies from 8–10 meV with 3 min interruption. The accompanying decrease of the luminescence intensity is undesirable and is probably due to accumulation of impurities during interruption. Indeed, on incorporation of a superlattice at the heterointerface instead of growth interruption, not only is the linewidth small (8–9 meV), but the peak PL intensity remains very high. We believe the superlattice traps impurities and smoothens the growth front.

The photoluminescence linewidth of the SQW is limited by the alloy quality in the well and

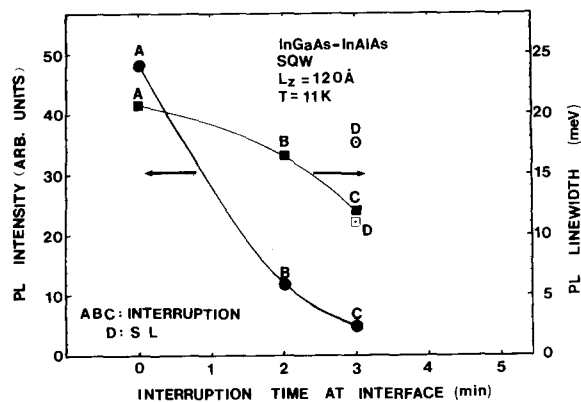


Fig. 1. Photoluminescence linewidth and intensity for a bound exciton peak in a 120 Å *InGaAs*–*AlInAs* quantum well. The points A, B and C represent cases where interruption time at each interface is 0, 2 and 3 min, respectively. D represents data for the case where a thin superlattice is placed before each interface without interruption.

barrier regions and the interface quality. Quantitative expressions have been derived by us earlier [3]. Furthermore, from theoretical studies of molecular beam epitaxial growth mechanisms [4] and from indirect studies based on RHEED measurements during growth [5], it appears that the normal interface may have a sharper profile compared to the inverted one. Thus, it is possible that the normal interface has a monolayer roughness. With this assumption, it is possible to estimate the parameters (height and lateral extent) characterizing the interface roughness. For example, analyzing the data obtained from the SQW grown with 3 min interruption, we conclude that the inverted interface has a roughness of the order of two monolayers while the lateral extent of the step is $\sim 100 \text{ \AA}$.

4. Properties of *In*_{0.52}*Al*_{0.48}*As*

From thermodynamic considerations one can determine whether or not an alloy can be produced without clustering. Due to the large difference between the In- and Al-related bond energies, the *InAlAs* system is expected to show clustering at the low temperatures employed in MBE growth. Previous work on liquid phase epitaxy suggests the presence of a miscibility gap in this alloy below 800°C [6]. However, since MBE is a far from equilibrium growth technique, in principle one may be able to grow cluster free alloys.

Computer simulation studies based on Monte Carlo techniques have revealed a basic incompatibility in the growth of cluster free alloy and atomically abrupt surfaces in conventional MBE growth when thermodynamic considerations favor clustering. For simplicity one may assume that the bulk alloy may be represented by two types of clusters with at least N_c atoms in the smaller-sized cluster and with concentrations C_A^1 and C_A^2 . In a perfect random alloy $N_c = 1$ and $C_A^1 = 1$ (0); $C_A^2 = 0$ (1). It can then be shown that the exciton linewidth σ is

$$\sigma = \sigma_0 n_c^{3/2} |C_A^1 - C_A^2|, \quad (1)$$

where σ_0 is the exciton linewidth when the alloy is random.

A similar model shows that the low-field mobility in the clustered alloy is given by

$$\mu^{-1} = \mu_0^{-1} n_c^3 |C_A^1 - C_A^2|^2, \quad (2)$$

where μ_0 is the mobility in absence of clustering. These expressions are valid if the spatial extent of the cluster is less than the exciton size in the alloy (~ 200 Å) and the electron wavelength. If the cluster sizes are larger, one expects to see multiple peaks in the PL spectra and a strong temperature dependence in the mobility.

A simple way to avoid clustering effects is to grow the material at very low temperature (e.g. 450°C for InAlAs) so that the cations have little migration after impingement. However, this produces a very poor quality of the growing surface and may result in the creation of native defects.

4.1. Photoluminescence measurements

Low temperature photoluminescence measurements on InAlAs indicate that the excitonic linewidth decreases with decrease of layer thickness, decrease of growth rate and increase of growth temperature. Fig. 2 shows the low temperature

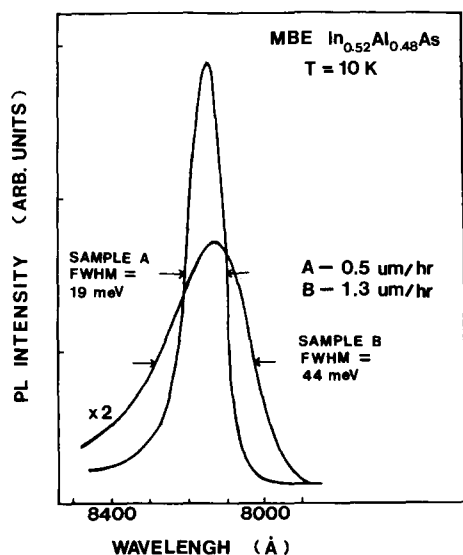


Fig. 2. Main bound exciton transition in low temperature photoluminescence spectra on *In*_{0.52}*Al*_{0.48}*As*. Case A and B represent growth rates of 0.5 and 1.5 $\mu\text{m/h}$, respectively.

excitonic peak for two samples grown at 490°C, but with different growth rates. It is clear that the linewidth is reduced by more than a factor of two when the growth rate is reduced from 1.3 to 0.5 $\mu\text{m/h}$. At the same time the peak intensity also increases. However, even a FWHM of 19 meV is much larger than the ideal linewidth of ~ 4 meV limited by alloy quality in these compounds. The larger linewidths can be explained either by the presence of a large density of defects or by the existence of clustering. The latter can be verified by X-ray and TEM studies.

4.2. Hall measurements

Since undoped InAlAs tends to be of high resistivity, measurements were made with Si-doped samples in which the room temperature electron density varied in the range $(1 \times 5) \times 10^{16} \text{ cm}^{-3}$. The mobility was strongly dependent on growth conditions and was higher in the samples grown at a slower rate. For a doping of $5 \times 10^{16} \text{ cm}^{-3}$, the mobility varied from 700 to 2200 $\text{cm}^2/\text{V} \cdot \text{s}$. Temperature-dependent Hall measurements were made to determine the compensation in the samples. We will, however, discuss here the behavior of the mobility for $T > 300$ K. It was observed that for samples in which the mobilities were low, the mobility increased with increase of temperature. This behavior is depicted in fig. 3. Compositional inhomogeneities can lead to this behavior and has been observed by one of us earlier [7] in the InGaAsP alloys, where also a miscibility gap is known to exist. Marsh [8] has explained this behavior by invoking alloy clustering and Mott and Davis [9] have predicted such increase in mobility in insulating materials due to transport by hopping conduction. The physical reason for the increase in mobility at high temperature can be understood as follows. The scattering rate due to alloy fluctuations are given by the matrix elements of the form

$$\int e^{i(k-k') \cdot r} v(r) d^3r,$$

where

$$v(r) = \begin{cases} v_0, & r < r_c, \\ 0, & r > r_c. \end{cases}$$

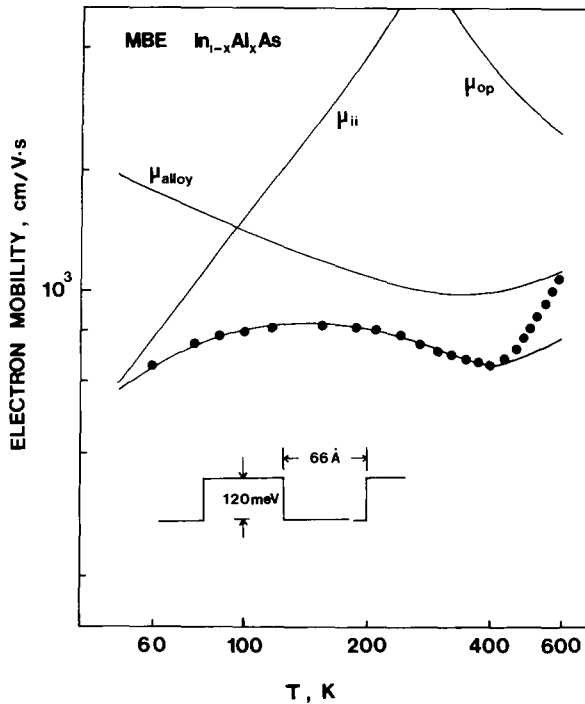


Fig. 3. Measurement and calculated temperature dependent Hall data in $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$. The inset shows the composition modulation resulting from alloy clustering.

The value of such matrix elements in general goes through a peak when $(k - k') \cdot r_c \sim 1$. Since $(k - k') \sim k_f$ after the k -space integration, one can roughly estimate the cluster sizes from the temperature at which the minimum in the mobility takes place. Using this analysis to fit the data, we estimate that $r_c \sim 33 \text{ \AA}$ and $|C_A^1 - C_A^2|$ is $\sim 4\%$. At very high temperatures, the discrepancy between theoretical and calculated mobilities is due to a sharp rise in the background electron concentration caused by ionization from a deep donor level (0.5 eV) and the resulting screening effects. Preliminary measurements of the impact ionization coefficients in the alloy give $\alpha/\beta = 4$. This result can be explained by considering a modulation of the threshold ionization energies due to clustering [10].

5. Studies on GaAs: In grown by MBE

We have investigated the effects of adding small amounts of In (0.2–1.2%) to GaAs on the densi-

ties of dominant electron traps and the peak intensities of defect induced transitions in the excitonic spectra of these materials. The decrease in density of four dominant traps M3, M4, M6 and M7, according to the nomenclature of Lang et al. [11], are shown in fig. 4. A sharp decrease is also observed in the peak intensity of the defect-induced excitonic transition g [12] at 1.509 eV in the 11 K photoluminescence spectra [13].

Our present data seem to suggest that the deep level electron traps are probably native defects or complexes involving native defects. We believe these defects can be Ga vacancies, since according to the expected growth kinetics such vacancies can be partially filled by In atoms, which have a surface migration rate three times that of Ga atoms at the growth temperature, thereby lowering the trap densities. This conclusion agrees with that of Skromme et al. [14]. Our observations in the photoluminescence data also strongly support the argument that the observed exciton transition g in our samples originate from native defects (possi-

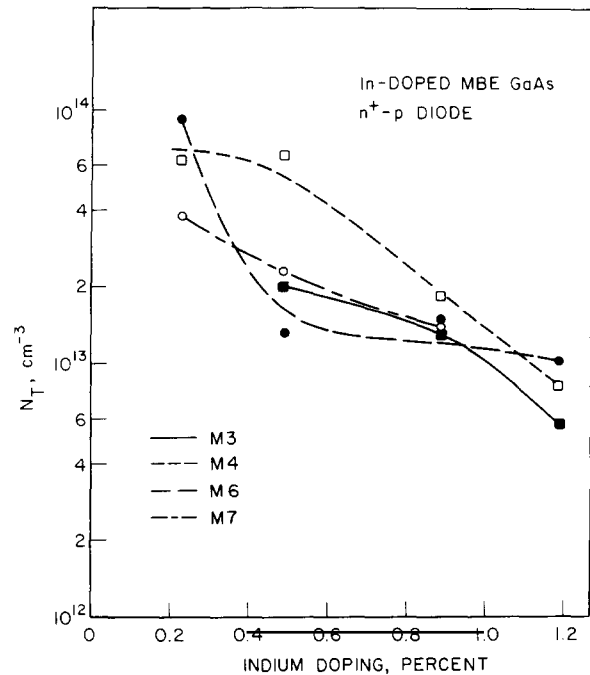


Fig. 4. Decrease in concentration of dominant electron traps with increasing In content in MBE grown GaAs.

bly V_{Ga}) or complexes involving native defects. This is in accordance with the hypothesis and observations of Briones and Collins [15] and Skromme et al. [14] that these emissions originate from complexes of native defects and C acceptors. It should be pointed out that the peak intensities of the free and donor- and acceptor-bound exciton transitions also decrease slightly upon addition of In. This result indicates that a reduction of dislocation density, as observed by Beneking et al. [16] in the case of LPE GaAs, is not a plausible mechanism for In-doped MBE GaAs. In the latter case a large increase of edge luminescence intensity was observed. The decrease observed in our samples may arise from impurities in the In, in spite of using the purest material commercially available.

It is known that the refractive index of GaAs is increased by the incorporation of small amounts of In in the lattice [15], which can be ideally used for light confinement. Practical waveguides using this scheme can either be homogeneous single epitaxial layers, in which case the In content needs to be small to avoid lattice mismatch effects, or strained-layer superlattices (SLS's) in which the $\text{In}_x\text{Ga}_{1-x}\text{As}$ well regions can have x as high as 0.5. We have recently fabricated ridge waveguides with losses as low as 1 dB/cm using these materials [17]. Coupled single-mode guides, 2 μm in width and separated by 1.5 μm were formed by photolithography and ion-milling. The lengths of the directional couplers varied from 0.5 to 2.5 mm. To effect electro-optic switching, the active guiding layer has on top of it a be-doped p^+ layer and an appropriate ohmic contact for the application of biases. 75% of the input light was directionally coupled between two such SLS guides having lengths ~ 1 mm. Efficient electro-optic switching on the application of a reverse bias of 4 V was also achieved. It should be mentioned that strong vertical confinement within the In-doped guiding layers is observed for these devices due to the incremental increase in refractive index caused by In. In this respect the devices are better than GaAs devices.

Acknowledgements

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