

# Formation of $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ heteroepitaxial layers using a pulsed laser driven rapid melt-solidification process

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Heteroepitaxial  $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$  structures have been formed for the first time by pulsed laser induced mixing of molecular beam epitaxy deposited In films ( $\sim 200 \text{ \AA}$ ) on GaAs (100) substrates. The process occurs by a melt-induced, rapid-mixing and solidification process driven by a XeCl pulsed excimer laser. The laser has a 27 ns full width at half maximum pulse width at 308 nm with its energy density of  $0.28\text{--}0.61 \text{ J cm}^{-2}$  homogenized into a  $4 \times 4 \text{ mm}$  square area which is stepped across the wafer.  $\text{In}_x\text{Ga}_{1-x}\text{As}$  layers with  $x$  values, as determined by both x-ray diffraction and Rutherford backscattering spectrometry simulation ranging from  $x = 0.21\text{--}0.26$  and thicknesses of 77–94 nm, have been formed. The formation of single-crystal layers has been verified by  $^4\text{He}$  ion channeling and cross-section transmission electron microscopy.

Alloy layers of  $\text{In}_x\text{Ga}_{1-x}\text{As}$  are being investigated by many research groups for a variety of potentially interesting applications. These include the use of strained  $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$  layers for heterostructure bipolar<sup>1</sup> and quantum well devices.<sup>2</sup> Also, another important area is the use of  $\text{In}_x\text{Ga}_{1-x}\text{As}$  for the fabrication of ohmic contacts to  $n$ -type GaAs.<sup>3–6</sup> The potential application here is to replace the conventional AuGeNi alloy ohmic contact to improve reproducibility, uniformity, and stability. To date,  $\text{In}_x\text{Ga}_{1-x}\text{As}$  layers are primarily grown using molecular beam epitaxy (MBE) techniques which require heating of the substrate. This severely limits the potential for *selective* growth on structures upon which devices already exist and is one factor limiting the development of integrated optoelectronic circuits containing  $\text{In}_x\text{Ga}_{1-x}\text{As}$  and GaAs devices.

In this letter we present initial results on a *laser-induced epitaxy* technique for the growth of heteroepitaxial  $\text{In}_x\text{Ga}_{1-x}\text{As}$  layers on GaAs. In this process pure amorphous In is deposited onto the clean surface of a GaAs (100) wafer and then intermixed using a rapid melt and recrystallization process driven by a pulsed XeCl excimer laser. The laser mixing produces uniform single-crystal layers of the alloy which exhibit excellent crystallinity. This process is similar to our previous work on the pulsed laser formation of epitaxial  $\text{Ge}_x\text{Si}_{1-x}$  layers on Si.<sup>7</sup> The In mole fraction and layer thickness are determined by the initial deposited In layer thickness and the laser power. As in our previous effort, the process is insensitive to minor interfacial contamination as it melts through the initial interface. Also, the interface abruptness can be controlled by the laser energy and number of pulses allowing one to grade the In mole fraction from the interface.

Earlier attempts<sup>8</sup> to produce  $\text{In}_x\text{Ga}_{1-x}\text{As}$  layers on

GaAs have employed continuous-wave (cw)  $\text{CO}_2$  and pulsed ruby lasers to react both deposited and implanted In layers on GaAs. The general outcome of this work was that it was difficult to form  $\text{In}_x\text{Ga}_{1-x}\text{As}$  heteroepitaxial layers. The problems with the cw laser is in the control of the energy deposition time; that is, it is very difficult to prevent over heating the area being reacted because the laser is scanned across the region and dwell times are generally  $> 1 \mu\text{s}$ . For both lasers selected, the areal energy deposition profile is quite nonuniform, being Gaussian for the case of the  $\text{CO}_2$ , and most likely highly spatially nonuniform for the ruby with significant hot spots remaining even after homogenization. A further problem also results from the deep energy absorption length for both wavelengths which results in significant nonuniform heating of the multilayer structure in depth.

The details of the processing in this work are as follows. The substrate is standard  $n^+$  (100) GaAs doped with Si. The wafer was cleaned first by organic solvents (acetone and methanol), then  $\text{H}_2\text{SO}_4:\text{H}_2\text{O}_2:\text{H}_2\text{O}$  (5:1:1) for 70 s,  $\text{NH}_4\text{OH}:\text{H}_2\text{O}_2:\text{H}_2\text{O}$  (1:1:10) for 60 s, and finally a de-ionized (DI) water rinse for 60 s. Prior to the In deposition, the GaAs oxide was desorbed at  $\sim 600^\circ\text{C}$  with the As source in a Varian GEN II MBE chamber in order to minimize the effects of oxygen on the layer. Reflected high-energy electron diffraction (RHEED) is used to monitor the desorption of the oxide, and the wafer showed  $2 \times 4$  RHEED reconstruction pattern after the desorption. The wafer is then cooled down to  $50^\circ\text{C}$ , and a 22-nm-thick layer of In was deposited onto the wafer from the MBE Knudson effusion cell at a rate of about 0.3 monolayers per second. The As beam was shut off when the wafer temperature reached  $400^\circ\text{C}$ . However, due to the configuration of this particular system, there was a residual As beam even when the As shutter was closed.

Following MBE deposition, the wafers are transferred to a pulsed laser system similar to that described previously

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for the gas immersion laser doping (GILD)<sup>9</sup> process. The sample is mounted in a cell with a quartz window. For this experiment argon gas was used in the cell, although, for *in situ* doping of the  $\text{In}_x\text{Ga}_{1-x}\text{As}$  layers, a suitable dopant gas can be included. Areas of the wafer are then irradiated by a XeCl excimer laser. This laser produces  $\sim 28$  ns full width at half maximum (FWHM) pulses of 308 nm light. The laser beam passes through an optical system which homogenizes the energy fluence to  $\pm 3\%$  over a  $4 \times 4$  mm square before striking the sample. The depth and  $x$  value of the layer are controlled by the energy fluence and number of pulses. Calibration of the laser is performed by melting a sample of pure Si. The melt duration, used as a process control variable, is measured by monitoring the transient reflectivity change of the layer using a HeNe laser probe.

Formation of the  $\text{In}_x\text{Ga}_{1-x}\text{As}$  layer occurs when the laser pulse melts the deposited In film and a variable thickness of the underlying GaAs substrate. The elements intermix in the liquid phase and regrow by fast liquid phase epitaxy on the (100) GaAs surface. The XeCl radiation is adsorbed in the near-surface region of the sample. Excess energy is delivered to this surface region and stored as specific heat. This energy propagates into the sample, melting it to a depth essentially proportional to the laser energy fluence and melt duration.

Rutherford backscattering spectrometry (RBS) is used to determine the alloy layer thickness, mole fraction  $x$ , and uniformity, while channeling alignment is used to determine the layer crystallinity. For this analysis we use 2.2 MeV  $^4\text{He}^+$  collimated to  $\sim 0.3$  mrad. The scattered particles are detected at angles of  $170^\circ$  and  $130^\circ$  relative to the incident beam. This allows us to specify the  $\text{In}_x\text{Ga}_{1-x}\text{As}$  layer thickness and composition in support of our x-ray diffraction and cross-sectional transmission electron microscope (XTEM) measurements.

Figure 1 shows  $170^\circ$  RBS random and aligned spectra for 220 Å In/GaAs (100) as-deposited and following ten laser pulses. This area is processed at an energy of  $0.61 \text{ J/cm}^2$

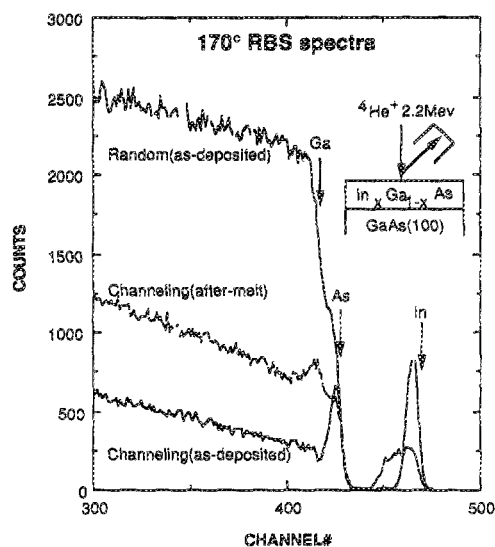


FIG. 1. RBS  $170^\circ$  random and  $\langle 100 \rangle$  channeling spectra of as-deposited and after laser melt samples. The energy density used for the laser-melted sample is  $0.61 \text{ J/cm}^2$  which is sample B in the following figures.

and intermixes to form an alloy layer of  $\text{In}_x\text{Ga}_{1-x}\text{As}$  which is  $\sim 900$  Å thick. The 220 Å amorphous layer on the GaAs substrate can be seen as the high-energy peak in the unprocessed aligned spectra. Following laser pulsing at an energy of  $0.61 \text{ J/cm}^2$ , the In has interdiffused into the GaAs substrate. The aligned spectrum for this condition is also shown in Fig. 1. Increased dechanneling is seen in the near-surface region indicating that either dislocations or lattice strain are present in the layer. The  $x$  value for this sample was determined independently by x-ray diffraction to be 0.22. For commensurate layer growth at this  $x$  value by conventional techniques,<sup>10</sup> the maximum thickness would be 200 Å. Because this layer is much thicker we expect to have misfit dislocations which could explain the increased dechanneling seen in the figure.

In Fig. 2 we show (a) the  $130^\circ$  RBS aligned In spectra and (b) their associated x-ray diffraction spectra. The laser energy fluence and number of pulses are indicated on the figure. Several trends can be observed from this figure. (i) For increasing laser energy fluence the  $\text{In}_x\text{Ga}_{1-x}\text{As}$  layer is both thicker, as seen by comparing the width of the RBS spectra for A versus B or C, and has a lower In mole fraction. (ii) For increasing numbers of pulses at a given thickness, the disorder peak at the layer-substrate interface decreases as seen by comparing the channeling results of B and C. This effect is most likely due to a changing In concentration profile in this region caused by the velocity dependence of the segregation coefficient of In in GaAs, i.e., the In profile at the melt interface has a concentration gradient which is a function of the number of laser pulses.<sup>11</sup> Using the RBS and x-ray diffraction data the  $x$  values and thicknesses of the films can be determined and are listed in Table I. The sharpness of the x-ray peaks indicates that the  $\text{In}_x\text{Ga}_{1-x}\text{As}$  layers are heteroepitaxial on the GaAs substrate.

In Fig. 3 we show cross-sectional bright field TEM images of the as-deposited and laser-processed samples A and C. The selected area diffraction (SAD) pattern, shown in Fig. 3(c), clearly shows that the  $\text{In}_x\text{Ga}_{1-x}\text{As}$  layers are

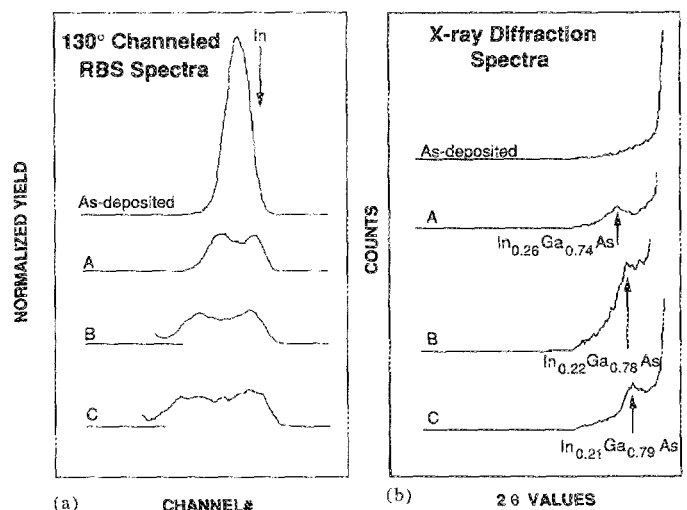


FIG. 2. (a) Aligned  $130^\circ$  RBS In peaks and (b) x-ray diffraction spectra of as-deposited and samples A, B, and C. The laser conditions used to process these samples are listed in Table I. The epitaxial layer thicknesses and compositions of samples A, B, and C are 770 Å  $\text{In}_{0.26}\text{Ga}_{0.74}\text{As}$ , 900 Å  $\text{In}_{0.22}\text{Ga}_{0.78}\text{As}$ , and 944 Å  $\text{In}_{0.21}\text{Ga}_{0.79}\text{As}$ , respectively.

TABLE I. Laser conditions used to process samples A, B, and C, and the corresponding layer compositions and thicknesses.

Sample No.	Laser energy (J/cm <sup>2</sup> )	Melt duration (ns)	Pulse No.	Composition	Layer thickness (Å)
A	0.53	76	50	In <sub>0.26</sub> Ga <sub>0.74</sub> As	770
B	0.61	92	10	In <sub>0.22</sub> Ga <sub>0.78</sub> As	900
C	0.61	119	40	In <sub>0.21</sub> Ga <sub>0.79</sub> As	940

crystalline on the GaAs substrate. Also, the composition calculated from these two patterns and the layer thickness estimated from the XTEM micrographs, are in good agreement with our RBS and x-ray diffraction measurements. In all of the calculations, the lattice parameters as a function of composition  $x$  in In <sub>$x$</sub> Ga <sub>$1-x$</sub> As needed for x-ray diffraction and SAD measurements were obtained by linearly extrapolating between the values for InAs and GaAs.<sup>12</sup> The uniformity of the In <sub>$x$</sub> Ga <sub>$1-x$</sub> As/GaAs interface is qualitatively quite good, although it is somewhat diffuse and hard to locate exactly. This we believe is due in part to grading in the composition at the interface caused by the velocity dependence of the segregation

coefficient of In in GaAs.<sup>11</sup> In comparing the XTEM images of samples A and C it can be seen that the lattice defect densities are significantly less in C than A. This is also seen in the increased spacing between the dark islands in the figure. It is possible that these islands are the source of the increased dechanneling seen in the RBS measurements of Figs. 1 and 2. The details of the nature of these islands are still under study. However, using energy-dispersive x-ray analysis (EDAX) and lattice imaging, we can definitely say that they are not associated with contaminant clusters or oxides occurring on the original deposited interface. The distribution and change in the shape (ellipse-like for sample A, and spherical for sample C) may provide insight into the rapid regrowth processes occurring.

In conclusion, pulsed laser mixing of deposited amorphous In layers on single-crystal GaAs (100) is shown for the first time to produce large-area, single-crystal In <sub>$x$</sub> Ga <sub>$1-x$</sub> As alloy layers. The absence of large numbers of dislocations provides evidence that the layers are commensurate with the GaAs substrate. Both the morphology of the layers and interfacial structure are excellent as evaluated by RBS and XTEM and HRXTEM. These exciting results are attributed to the correct choice of laser conditions, careful homogenization of the energy fluence, and correct choice of wavelength. Future work will continue the evaluation of the layers with emphasis on producing thinner In <sub>$x$</sub> Ga <sub>$1-x$</sub> As layers and the examination of strain in these layers. Continued investigation of the unusual defect found at the melt-solid interface will be pursued in order to shed light on the crystallization mechanism, particularly since they appear in the region of rapid acceleration of the liquid/solid interface during recrystallization.

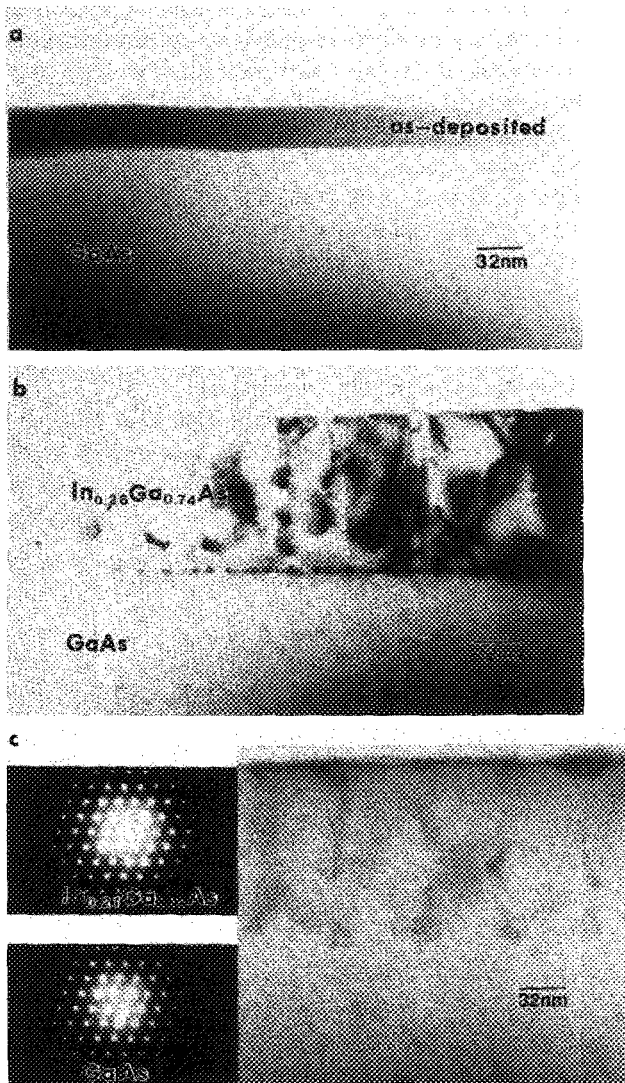


FIG. 3. Cross-sectional bright field transmission electron micrographs of (a) as-deposited, (b) sample A, and (c) sample C. The selected area diffraction patterns shown in (c) correspond to In<sub>0.21</sub>Ga<sub>0.79</sub>As and GaAs, respectively.

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