A study of strain-related effects in the molecular-beam epitaxy growth of $In_xGa_{1-x}As$ on GaAs using reflection high-energy electron diffraction

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In this paper we examine the role of strain in the molecular-beam epitaxy (MBE) growth process by studying growth in $In_xGa_{1-x}As$ on GaAs with x varying from 0 to 0.5. This range covers the critical thickness regime for dislocation formation from ∞ to 12 monolayers. We have studied MBE growth for both on-axis (100) and misoriented substrates. The first issue we address in this paper is the role of strain in controlling the atomic-surface migration. We find from reflection high-energy electron diffraction (RHEED) studies during growth of $In_xGa_{1-x}As$ that as x is increased, the surface migration decreases rapidly. The growth front of the growing structure roughens due to this decreased migration and we have studied the recovery time for the growth front to smoothen. The surface recovery time increases rapidly as the strain in the system increases. Conversely, when GaAs growth is resumed, there is a recovery of the RHEED average intensity and oscillations (peak to peak). At higher growth temperatures, layer-by-layer growth is again restored. Dynamic changes in the RHEED pattern were video recorded, and the data indicate that in the three-dimensional island growth mode observed, the lattice constant in the strained overlayer changes monotonically and attains an equilibrium value before the conventionally calculated critical thickness h_c is reached. Low-temperature photoluminescence measurements on misoriented (0°–4°) InGaAs/InAlAs MQW grown directly on GaAs indicate that device-quality material can be obtained for growth at high temperatures (~ 570 °C).

I. INTRODUCTION

The desire to tailor electronic and optical properties of semiconductors for specific device applications has led to a considerable research activity in the area of strained epitaxy.[1,2] A great deal of research has also focused on growth of strained structures not only for material tailoring, but for applications in the area of integration of optical and electronic devices.[3-6] The critical growth issues for these two aspects, namely, the material tailorability and integration are quite different. For the former applications, it is desirable to avoid any dislocations, a requirement that forces one to keep the epilayer thickness smaller than the critical thickness.[3] For latter applications, however, one often needs thick layers (several microns) and the important requirement is to produce dislocations to accommodate strain. However, once the dislocations are produced, the quality of the grown layer depends upon ensuring that the dislocations do not propagate in the growth direction. Using the terminology commonly employed to describe such dislocations, it is preferred to have misfit dislocations and to avoid screw or threading dislocations.

Reflection high-energy electron diffraction (RHEED) is an important *in situ* analysis technique which is capable of giving semiquantitative and quantitative information about the growth process and its control.[7-10] A considerable amount of insight into the growth process has also been gained from computer simulation studies.[11,12] However, since there are a number of important unknown parameters used in the simulation models, it becomes necessary to estimate these from RHEED data.

The motivation of the present investigations was twofold. First, the role of strain in the surface migration of impinging atoms and consequently on the quality of the growing surface needs to be understood. Second, the accommodation of strain in the growing overlayer is not well-understood at present, although some models have been proposed which are used for calculations of the critical thickness and other related parameters. In particular, there is a need to understand the changes in the lattice as one grows a coherently strained layer and beyond this regime to the region where dislocations are produced. In the present study, we have examined the role of the molecular-beam epitaxial (MBE) growth process of $In_xGa_{1-x}As$/GaAs with x varying from 0 to 0.5. This range covers the critical thickness regime for dislocation formation from ∞ to approximately 12 monolayers (ML). We have studied MBE growth on both on-axis (100) and misoriented substrates.

II. THEORETICAL CONSIDERATIONS

It is important to discuss the physical nature of MBE growth and the information provided by the RHEED ex-

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periments. Much of the understanding of the MBE growth process has evolved from the work of Arthur[13] and Foxon and Joyce[14] on GaAs. Computer simulations based on Monte Carlo methods have also provided a great deal of insight into the atomistic nature of MBE growth.[11,12] By a combination of these experimental and theoretical techniques, it has been demonstrated that the growing crystal will retain a smooth and atomically abrupt surface only if the cation surface migration rate is very high ($\geqslant 10^4$ hops/s). If the average length traveled by cations is larger than a step edge on a misoriented substrate, the growth occurs by a layer-by-layer mode.

The cation surface migration rate can be expressed as

$$R^{i}_{\parallel} = R_{\mathrm{od}} \exp(- E^{i}_{\mathrm{tot}} - E_{\parallel}/kT_s) \qquad (1)$$

for in-plane hops and

$$R^{i}_{\perp} = R_{\mathrm{od}} \exp(- E^{i}_{\mathrm{tot}} - E_{\perp}/kT_s) \qquad (2)$$

for interlayer hops. R_{od} is a prefactor which has negligible temperature dependence. E^{i}_{tot} is the total energy with which the cation at site i is bonded to the growing crystal and $E_{\parallel}$ and $E_{\perp}$ are the minimum energies with which the cation is bonded on the surface as it hops from site i to the next site. It is clear that the hopping rate is controlled by the surface bond strengths of the cations. In strained systems, the surface bond strength is expected to be affected by the local strain so that the migration rate may be quite different. However, at present there is no understanding of how this occurs. Information on this may be provided by RHEED intensity oscillations. The RHEED intensity from a growing surface is given by

$$I = \left| \sum_j \exp[i(k - k')\cdot r_j] \right|^2, \qquad (3)$$

where k is the momentum of the incident electrons, which form a collimated beam, and k' is the momentum in the direction of the detector. The summation in Eq. (3) is restricted to the surface atomic sites since the de Broglie wavelength for the electrons is ~ 0.1 Å. It is easy to see that away from the Bragg angle, the intensity from surface atoms on successive monolayers will interfere destructively. Thus, under conditions in which growth is taking place in a layer-by-layer mode (so that the cation migration is high) and the surface profile is changing (e.g., if one is growing on axis with no surface steps), one should expect the RHEED intensity to oscillate. These oscillations will eventually die out because of the increasing surface roughness and because of flux nonuniformities. If the flux nonuniformities are negligible, then the magnitude of the oscillation is representative of the quality of the growing front. We note, as has been pointed out by several workers,[3] that one may not see any oscillations on stepped surfaces even though one may be growing under ideal growth conditions.

Theoretical understanding of strained epitaxy under nonequilibrium conditions is still in its infancy, even though the problem was initially addressed nearly 40 years ago. A number of workers have studied the problem of strained structure and established the criteria for the formation of dislocations.[15-17] These studies are based on obtaining the minimum energy state between the overlayer and the substrate using analytical, numerical, and recently, Monte Carlo techniques. The results of these energy minimization calculations can be summarized in general as follows:

For a single monolayer of one- or two-dimensional strained overlayer with lattice mismatch Δa one can define three regions. (i) $\Delta a \leqslant \Delta a_{c1}$, where no dislocations are produced and the strain is absorbed coherently. (ii) $\Delta a_{c1} \leqslant \Delta a \leqslant \Delta a_{c2}$, the lowest energy state is one containing dislocations, but metastable states containing no dislocations may be obtained. (iii) $\Delta a_{c2} \leqslant \Delta a$, dislocations are always produced.

In three-dimensional strained layer overgrowth, one may again define a critical thickness h_c for every Δa, where below h_c, the lowest energy state has no dislocations, while above h_c, dislocations are produced. Once again, metastable states with no dislocations can be produced for thicknesses above h_c. However, these studies have not addressed the important question of how dislocations arise during growth and how their nature is controlled by the mode in which the crystal is growing. This is a difficult problem which most probably does not permit an analytical solution. A detailed experimental study of strain accommodation will thus provide a tremendous input to better theoretical understanding.

III. EXPERIMENTAL TECHNIQUES

MBE growth was performed in a three-chamber RIBER 2300 system. Strained InGaAs layers were grown on undoped-GaAs substrates. The substrates were initially solvent degreased. Mechanical damage resulting from polishing was removed by etching (5:1:1) (H_2SO_4:H_2O:H_2O_2). Surface oxides on the substrates were removed by a quick etch in

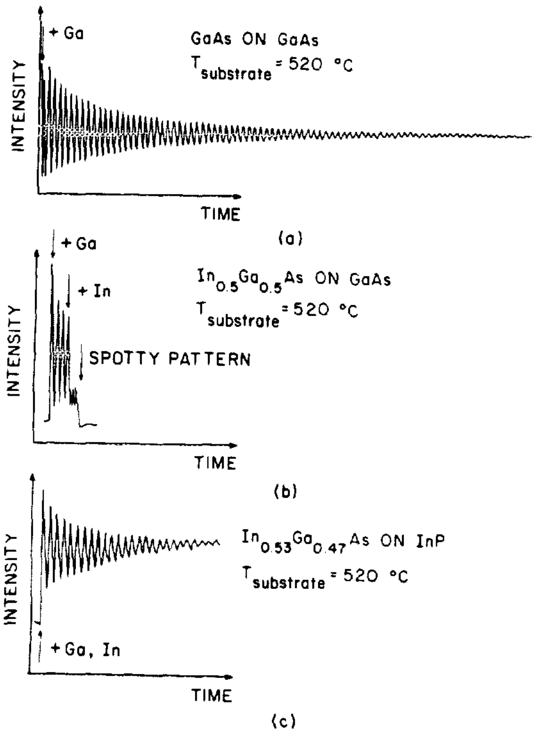

FIG. 1. Typical RHEED oscillations observed for growth of: (a) GaAs on GaAs; (b) $In_{0.5}Ga_{0.5}As$; and (c) $In_{0.53}Ga_{0.47}As$ on InP substrates at a growth temperature of 520 °C under As_4 conditions.

(1:1) (HCl:H_2O). The substrates were then rinsed in deionized water and amounted on the molybdenum blocks with indium. Prior to initiation of growth, oxides were desorbed at 625 °C under an As_4 flux. RHEED was used to monitor desorption of the oxides. Appropriate undoped-GaAs buffer layers 0.25 μm thick were grown prior to RHEED oscillation measurements. Arsenic to group III flux ratio of 80:1 was generally maintained during growth.

The RHEED system consists of a commercial 10 keV electron gun focused onto a phosphor-coated screen. Light from the diffraction patterns was collected by a lens/pinhole assembly mounted onto an *X–Y–Z* micrometer stage and focused onto a photomultiplier tube. The detected signal was suitably amplified and recorded. In order to observe the dynamic changes in the lattice constant during the growth of a strained layer in the coherent and dislocation regimes, a Panasonic video camera, sensitive to low illumination levels, was used for recording.

IV. RESULTS AND DISCUSSION

In Fig. 1, we show RHEED oscillation data for growth of GaAs on GaAs, $In_{0.5}Ga_{0.5}As$ on GaAs and for comparison, $In_{0.53}Ga_{0.47}As$ on InP,[18] all recorded for a substrate temperature of 520 °C and with As_4-stabilized conditions. The first and the last cases are for lattice-matched systems and show a long temporal persistence of the oscillations. On the other hand, the growth of $In_{0.5}Ga_{0.5}As$ on GaAs, characterized by a mismatch of 3.5%, shows strongly damped oscillations. The data indicate that increased compressive strain inhibits cation migration and thereby causes the growth front to roughen. It is possible that in the strained InGaAs, indium atoms assume a bonding configuration similar to that in GaAs, thereby possibly enhancing the surface migration activation barrier. This can lead to decreased surface mobility and increased roughness of the growth front. The observed decrease of the average RHEED intensity is a result of this roughness. This must be contrasted with the opposite expectation, since normally for unstrained systems the addition of In is expected to increase the average migration rate due to the weaker In–As bond.

Figure 2 shows the smoothness recovery time after growth interruption under As_4 flux for various alloy compositions with increasing In, up to 50%. In this series of experiments, we started growth of $In_xGa_{1-x}As$ on a smooth surface and measured the initial highest intensity of the RHEED oscillation I_0. The growth was then continued until the oscillations disappeared. We then interrupted growth by varying time periods t_i. The growth of $In_xGa_{1-x}As$ was then reinitiated and the initial intensity of the RHEED oscillation $I_i(t_i)$ was measured and compared with I_0. As the interruption time is increased, the two intensities approach each other signifying a better recovery with longer interruption time. The data support our observations in that the increased strain inhibits cation migration. It is clear that strain plays a very important role in the surface kinetics and hence in the growth mode and, therefore, may have considerable influence over the ideal growth conditions. Growth of high quality strained structures may require use of novel growth techniques to overcome this additional strain-related problem.

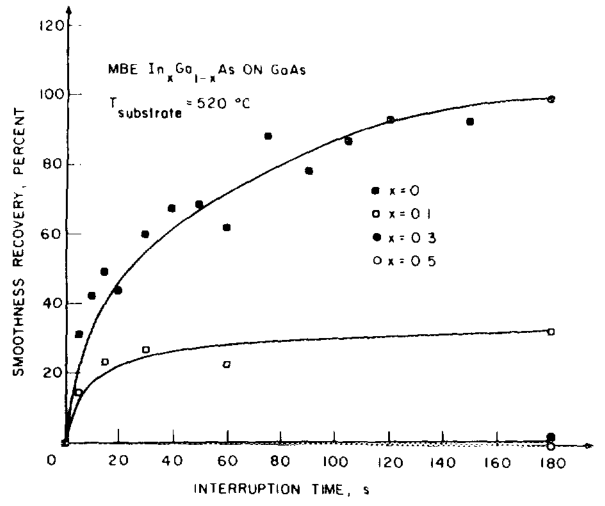

FIG. 2. Smoothness recovery vs interruption time during growth of $In_xGa_{1-x}As$ on GaAs at 520 °C for $x = 0.0$, 0.1, 0.3, and 0.5.

Figure 3 shows RHEED oscillation data during InGaAs/GaAs heterostructure growth. GaAs–$In_xGa_{1-x}As$–GaAs structures were growth with $x = 0.1$, 0.2, 0.3, and 0.5. A 0.25 μm GaAs buffer layer was first grown at 605 °C, after which the growth temperature was stabilized at 520 °C. GaAs growth was initiated again and upon completion of a few monolayers, the In flux was initiated. Several monolayers of InGaAs were grown and then the In flux was stopped again. The recovery of the pattern during GaAs growth was recorded. The two distinct features of the data are (a) the

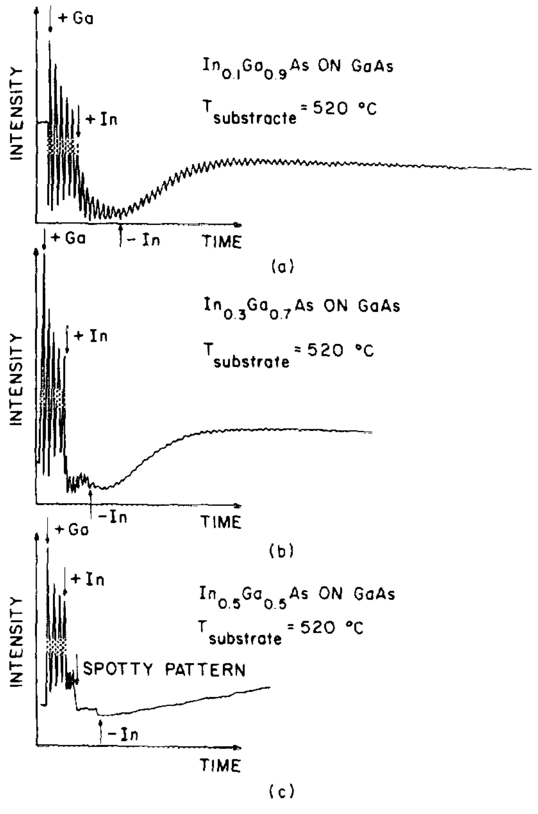

FIG. 3. RHEED oscillation data showing changes in oscillation intensity and average intensity during growth of GaAs–$In_xGa_{1-x}As$–GaAs heterostructures.

average RHEED intensity falls during growth of InGaAs, as observed in Fig. 1, and increased again during subsequent GaAs growth, and (b) as is clear from the data of $In_{0.1}Ga_{0.9}As/GaAs$, the magnitude of the RHEED oscillation also recovers during GaAs growth. We speculate that both phenomena have the same origin and are related to three-dimensional type growth. The change in RHEED intensity probably reflects the random change in lattice constant on these surface islands. The ratio of the fall and rise of the RHEED intensity will be analyzed in detail to be able to better understand the complexity in the growth modes during strained layer epitaxy.

In Fig. 4, we show the results for the average surface lattice constant of the growing strained overlayer as a function of thickness. This information is obtained from the video recording and analyzing the relative change in the spacing of the RHEED spots. We find that the lattice constant adjusts continuously from the GaAs lattice constant to the $In_xGa_{1-x}As$ lattice constant over a distance d_c, which is smaller than the calculated critical thickness h_c. This result is somewhat different from what is expected from the conventional understanding, that the lattice constant should remain equal to the substrate during coherent growth ($\leqslant h_c$) and change over to the overlayer lattice constant above h_c.[3]

It was observed that at growth at 520 °C, the pattern turns spotty, representing a three-dimensional island growth mode after a few monolayers. This is indicated in Fig. 1(b). Comparing this with Fig. 1(c), the above results indicate that while lattice-matched InGaAs/InP grows with atomically smooth surfaces, the presence of strain greatly inhibits surface mobility and introduces roughness. In another series of experiments, the strained layers were grown at much higher temperatures, approximately 575 °C. The pattern remained streaked for growth of a 0.05 μm film. The adatom mobilities are enhanced at this higher growth temperature, thereby restoring the smoothness.

The RHEED oscillations are very sensitive to the orientation of the substrate. In misoriented growth, the growth occurs on a stepped surface and if the average atomic migration length is larger than the step size, growth can occur in a mode where the surface profile does not change with time so that RHEED oscillations cannot be observed. As a preliminary study of misoriented growth, we have grown $In_xGa_{1-x}As$ on misoriented GaAs under conditions similar to the RHEED experiments. $In_xGa_{1-x}As/In_xAl_{1-x}As$ MQW structures were grown on oriented (100) and misoriented [2° and 4° off towards (011)] GaAs as follows: a 0.2 μm GaAs buffer layer was first grown at 610 °C followed by 250 Å of $In_xGa_{1-x}As$ ($0 \leqslant x \leqslant 0.3$) at 520 °C at a rate of 0.1 μm/hr. Growth was then interrupted and the growth temperature and rate were changed to 560 °C and 1 μm/hr, respectively. One micrometer of $In_xGa_{1-x}As$ followed by 10 periods of $In_{0.15}Ga_{0.85}As/In_{0.15}Al_{0.85}As$ MQW ($L_Z = L_B = 100$ Å) were grown continuously under these conditions. The morphology for the (100)-oriented and misoriented layers show a surface pattern (with spacing of approximately a few micrometers) related to misfit dislocations. Low temperature photoluminescence (PL) spectra measured in these samples are shown in Figs. 5 (a)–5 (c). The highest energy peak in the spectrum originates from quantum well-bound exciton emissions. The peak at lower energies originates from the bulk $In_{0.15}Ga_{0.85}As$ layer. A steady increase in the intensity of the MQW is observed with increase of misorientation. The integrated PL intensity increases by almost a factor of 2 in going from (100) to 4° off toward (011). These results confirm the physical picture of the role of misorientation and indicate that these misoriented heterostructures may be suitable for device fabrication.

V. CONCLUSIONS

In this paper, we have addressed some of the important issues in growth of strained layers. It is somewhat surprising

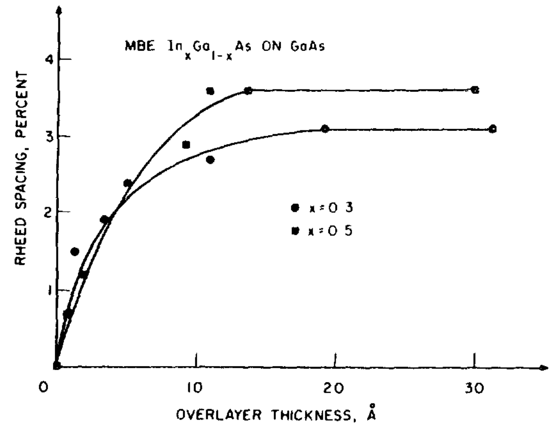

FIG. 4. Change in the separation between diffraction streaks on RHEED screen as function of overlayer thickness during growth of $In_xGa_{1-x}As$ on GaAs at 520 °C.

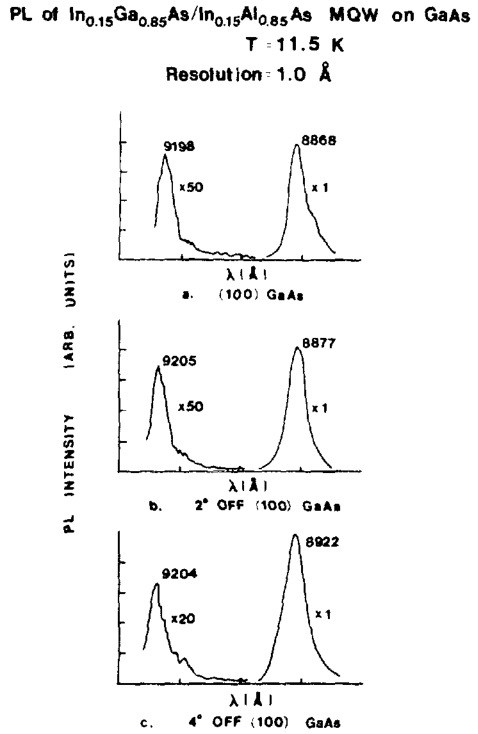

FIG. 5. Low temperature photoluminescence spectra of $In_{0.15}Ga_{0.85}As/In_{0.15}Al_{0.85}As$ grown directly on GaAs for (a) (100) orientation; (b) 2° off (100) towards (011); and (c) 4° off (100) towards (011).

to find that the introduction of strain reduces the atomic surface migration. Since atomic migration controls the quality of the growing surface, it is clear that optimum conditions for growth of lattice-matched $In_{0.53}Ga_{0.47}As$ on InP and $In_{0.5}Ga_{0.5}As$ on GaAs are very different. At present, it is not clear what these optimum conditions are and how they change with strain. Results obtained for higher temperature growth suggest that for layer-by-layer growth of strained systems, one requires a slow growth rate and/or a higher growth temperature. We are presently investigating the details of the variation of growth modes with increased substrate temperature.

We have also examined the variation of the surface lattice constant as a function of the strained overlayer thickness. We find a continuous change in the surface lattice constant as the thickness increases. For the three-dimensional island growth observed by us, the surface lattice constant approaches the bulk lattice constant of the overlayer *before* the calculated critical thickness. Further studies of the strained system especially with misoriented growth are continuing and may shed more light on the important issue of strained epitaxy.

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