Role of strain and growth conditions on the growth front profile of $In_xGa_{1-x}As$ on GaAs during the pseudomorphic growth regime

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Theoretical and experimental studies are presented to understand the initial stages of growth of InGaAs on GaAs. Thermodynamic considerations show that, as strain increases, the free-energy minimum surface of the epilayer is not atomically flat, but three-dimensional in form. Since by altering growth conditions the strained epilayer can be grown near equilibrium or far from equilibrium, the effect of strain on growth modes can be studied. *In situ* reflection high-energy electron diffraction studies are carried out to study the growth modes and surface lattice spacing before the onset of dislocations. The surface lattice constant does not change abruptly from that of the substrate to that of the epilayer at the critical thickness, but changes monotonically. These observations are consistent with the simple thermodynamic considerations presented.

Interest in strained molecular beam epitaxy (MBE) has been growing rapidly recently, and a number of groups have demonstrated high quality device results in systems which directly or indirectly involve strained epitaxy.[1-4] Devices where active regions are under coherent strain (pseudomorphic regions) are being used for high-speed electronic devices as well as optical devices.[1,2] In addition, material systems with incoherently strained regions (involving dislocation generation) hold tremendous potential for integrated technologies (GaAs/Si,CdTe/GaAs, etc).[3,4] In both these applications, the initial stages of growth (before dislocation generation or below "critical thickness") need to be understood if the full potential of strained epitaxy is to be realized. This understanding should be at an atomic level because in many pseudomorphic devices, device performance is sensitive to atomistic distance fluctuations.

In lattice-matched systems, the general nature of growth in MBE is fairly well understood (although a number of details are not). The growth front during MBE can be described as being in between two extreme growth modes: (a) a layer by layer growth mode where the growth front essentially consists of at most two exposed (cation) monolayers and even though atoms impinge randomly from the vapor phase, they move rapidly on the exposed surface and attach themselves (get incorporated) at step edges and (b) a three-dimensional growth mode where the impinging atoms have essentially no surface kinetics and the surface is rough due to the intrinsic statistical fluctuations. These growth modes can be readily observed by *in situ* reflection high-energy electron diffraction (RHEED). The above picture is a good description of the lattice-matched systems if the free-energy minimum for the growth surface favors an atomically smooth surface. For the (100) surface, which we will focus on in this letter, one has cation or anion layers and energetically the difference between a perfectly smooth [Fig. 1(a)] and a rough [Fig. 1(b)] surface is essentially due to in-plane bonds which are second neighbor bonds. If the second neigh-

bor bond strength was zero, the rough surface would be the lower free-energy surface (due to entropy considerations). The second neighbor bonds are expected to be an order of magnitude smaller than nearest neighbor bonds, but although small, at usual MBE temperatures, $|kT| < |W_2|$ (= second neighbor bond energy), and one has the possibility of a two-dimensional layer by layer growth mode.

In the case of strained epitaxy, in addition to considerations arising from second neighbor bond strengths, one has to also contend with the strain energy. The problem of strained layers under equilibrium has been studied for several decades,[5,6] although only recently the predictions on critical thicknesses in MBE grown III-V systems have been verified.[7-9] According to these theories (based on energy balance between strain energy and chemical energy due to dislocation generation) for any lattice mismatch ΔR, as long as the epilayer grown on a thick substrate is below a certain
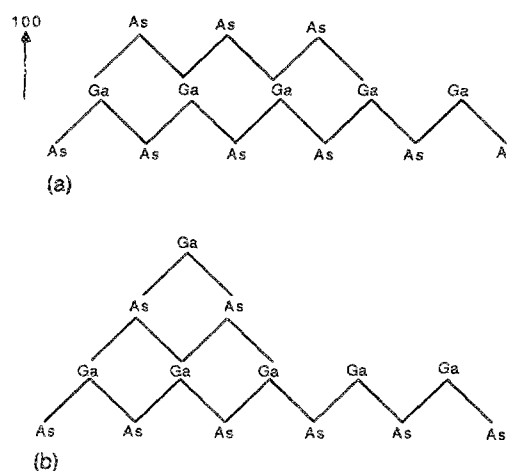

(a)

(b)

FIG. 1. (a) Atomically abrupt and (b) rough (100) surface. The atoms on the rough surface have the same number of nearest neighbor bonds but fewer second neighbor bonds.

thickness (the critical thickness), the strain is absorbed coherently and no dislocations are produced. However, above the critical thickness, misfit dislocations are produced at the epilayer-substrate interface. While such an assertion has been verified experimentally in a number of systems,[8,9] it is not clear how strain affects the growth mode below critical thickness. The simple arguments presented below show that the minimum free-energy surface in a strained system is not atomically smooth but has a three-dimensional (3D) form, with the bond lengths in the 3D islands corresponding to the free-epilayer bond lengths.

We will attempt to compare the energies of the atoms in a (100) surface when they are occupying a complete monolayer (atomically flat surface) and when they are arranged in three-dimensional islands. Let ΔR be the lattice mismatch and R_0 the substrate lattice constant. The average spacing between misfit dislocation arrays produced after critical thickness is achieved is $L_d \cong R_0^2 / \Delta R$. Let us divide the growth surface into squares of sides L_d as shown in Fig. 2. If all the atoms are arranged in the same monolayer, the average strain energy for all atoms in any of the squares is

$$E_s(L_d) \sim 4 \frac{L_d W_1}{d_c/R_0}, \qquad (1)$$

where the numerator represents the dislocation energy (W_1 = nearest neighbor bond energy), R_0 the monolayer distance, and d_c is the critical thickness. This equation is obtained from the fact that at critical thickness the total strain energy in the d_c/R_0 monolayers is equal to the dislocation energy. Let us now consider the case shown in Fig. 2(b), where the atoms instead of being in the same monolayer are forming simple three-dimensional islands of height n^2 and that there is no strain energy in the islands (the lattice constant in the islands is the same as in the bulk epilayer material).

For the first monolayer, the excess energy due to the substrate epilayer chemical energy is

$$E_{\text{sub}} \sim 4 \frac{L_d W_1}{n^2 d_c/R_0}. \qquad (2)$$

The factor n^2 comes since the number of the first monolayer atoms is reduced by n^2 in the case considered in Fig. 2(b). In addition, for the case of Fig. 2(b), there is also a change in energy because the edge atoms in the three-dimensional islands do not have second neighbor bonds. This excess chemical energy is

$$E_{\text{chem}} \cong n^2 4 (L_d/n) W_2 \qquad (3)$$

where n^2 is the number of monolayers in the island and W_2 is the second neighbor interaction.

Minimizing the energy difference between Eq. (1) and the sum of Eqs. (2) and (3) gives

$$n^3 \cong 2 (W_1/W_2)(R_0/d_c). \qquad (4)$$

Since $W_1/W_2 \sim 10\text{--}15$, this equation suggests that if $d_c < 20$ monolayers (i.e., strain > 2%) the strained epilayer will minimize its energy by forming a three-dimensional island surface. For growth of InGaAs on GaAs this would be expected for In composition greater than 25–30%. We note that in the above arguments we considered the two extreme cases of the atoms being arranged either coherently strained in a monolayer with the surface lattice constant the same as the substrate's or being arranged in little islands with unstrained lattice constant. In reality, the free-energy minimum surface will be between these two extreme cases. Nevertheless, the simple physical picture shows that the presence of strain forces the system to go towards a 3D island mode grown for conditions near equilibrium. The surface lattice constant for Fig. 2(a) should be the same as that of the substrate lattice constant until the critical thickness and then abruptly change to the epilayer bulk lattice constant. On the other hand, for Fig. 2(b) the surface constant should show a monotonic change from the substrate to the bulk epilayer value.

We have carried out a series of experiments involving growth of InGaAs on GaAs under various growth conditions and used *in situ* RHEED studies (oscillations and spacings) to monitor the growth front. GaAs substrates were cleaned normally and oxides were desorbed under arsenic overpressure. GaAs buffer layers of thickness 0.2 μm were first grown at 610 °C at a rate of 0.39 μm/h. Growth was then interrupted, the substrate temperature was suitably altered and the 2×4 reconstruction pattern was displayed on the screen. Growth was resumed by growing 1000 Å of $\text{In}_{0.35}\text{Ga}_{0.65}\text{As}$ with a 5 s delay in opening the In shutter, while videotaping the RHEED pattern with a Panasonic system sensitive to low illumination levels. The growth of the ternary was done at several substrate temperatures and at the growth rate of 0.6 μm/h.

In Fig. 3(a) we show the initial states of RHEED oscillations as growth of InGaAs is initiated on an on-axis smooth GaAs. Note the extremely rapid decay of the oscillations clearly suggesting that while in the lattice matched region of the growth (GaAs on GaAs) the RHEED oscillations show a layer by layer growth, in the strained case the
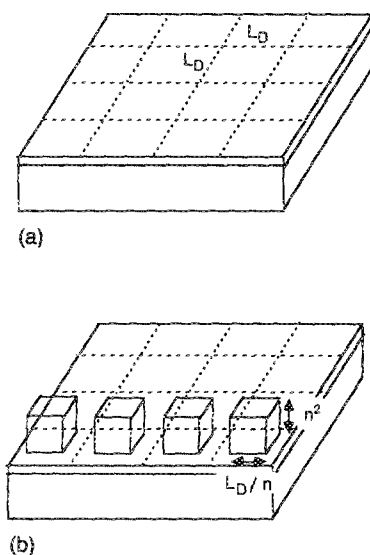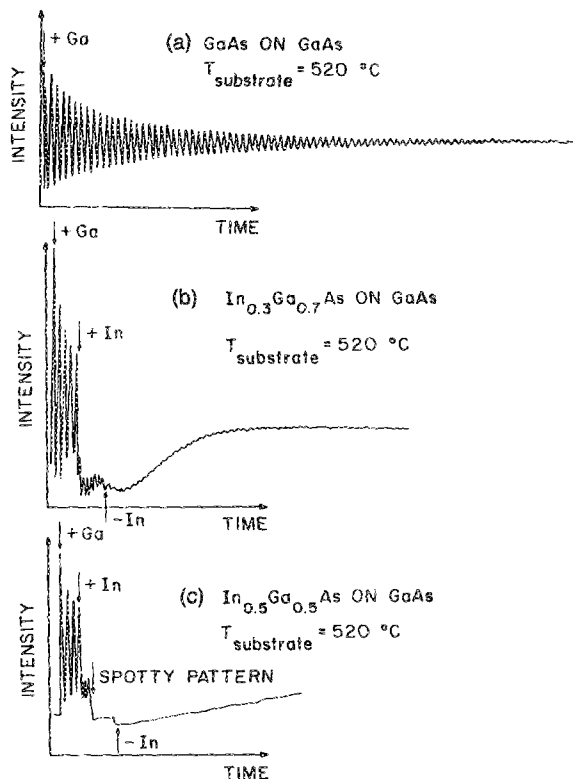

(a)

(b)

FIG. 2. Schematic showing two cases (a) where a monolayer of the lattice mismatched epilayer is arranged in an atomically flat monolayer under coherent strain and (b) where the atoms form 3D islands with bases of side L_d/n and height n^2.

FIG. 3. RHEED oscillation data for growth of (a) GaAs on GaAs, (b) $In_{0.3}Ga_{0.7}As$ on GaAs, and (c) $In_{0.5}Ga_{0.5}As$ on GaAs.

FIG. 4. Surface lattice constant measured by RHEED integral order spacing with increasing overlayer thickness of MBE grown $In_{0.35}Ga_{0.65}As$ on GaAs at various substrate temperatures. The arrow represents the calculated critical thickness according to the formulation of Ball and Van der Merwe (see Ref. 5).

growth mode abruptly changes to a 3D island mode. We also see that as the strain (In composition) is increased, the transition to the 3D mode is more abrupt. Note that in all cases shown the epilayer thickness is below the critical thickness, consistent with the theoretical arguments presented above, and suggests that the close-to-equilibrium surface for the strained epilayer is indeed 3D in nature.

In Fig. 4 we show the change in surface lattice constants as obtained from the RHEED integral order spacings. Data are obtained by digitizing individual frames of the videotape into a 480×480 array with 256 grey scale and loading into the computer. Slices through the array were taken across the integral order RHEED lines and the intensity distribution plotted. By analysis of the dynamic changes of these peaks, we see dramatic movement at higher temperatures. At low temperatures, where impinging atoms are unable to move in a correlated manner to reach the free-energy minimum surface, the lattice constant remains close to the substrate's. However, at higher temperatures as the surface kinetics is increased, the monotonic change of the lattice constant expected from the simple energy balance treatment is indeed observed. It should be mentioned that similar data have recently been presented by Whaley and Cohen.[10]
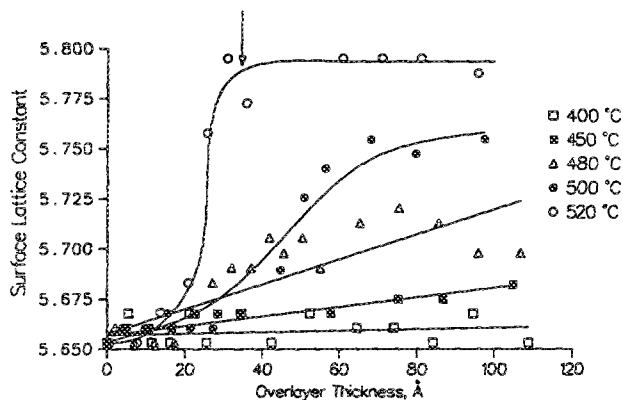
In summary, we have presented theoretical and experimental results on the initial stages of growth (before dislocation generation) of InGaAs on GaAs. It is clear that strain plays a key role in controlling the growth mode and that the growth condition dependence of growth modes of lattice matched and strained epilayers are quite different.

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