Critical-Thickness and Growth-Mode Transitions in Highly Strained $In_xGa_{1-x}As$ Films

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Singular phenomena in highly strained $In_xGa_{1-x}As$ are used to test current theories of dislocation dynamics in thin films. Strong support is found for a temperature-dependent frictional force which has an activation energy of magnitude of the Peierls energy. With this force, an abrupt temperature-dependent transition in the critical thickness of pseudomorphic growth is explained for the first time; the island-to-layer growth-mode transition which occurs at this temperature is shown to be equivalent to a similar x-dependent transition; and island growth is attributed to nucleation by misfit dislocations.

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Strained-layer semiconductors are complex materials and are not presently well understood. For the growth of InGaAs on GaAs at substrate temperatures near 530 °C, there is some agreement between experiment and a simple temperature-independent theory of critical thickness (the thickness beyond which the InGaAs ceases to grow pseudomorphically) over a range of indium concentrations.[1,2] At lower growth temperature the experimental observations are not adequately explained and in particular, the role of time-dependent plasticity is unclear.[3-5] Matthews and co-workers extended the simple theory with a velocity- and temperature-dependent glide activation force[6,7]—referred to as a frictional force. Dodson and Tsao[8,9] introduced the concept of "excess stress" in analogy with plasticity theory for bulk semiconductors.[10] More recently Fox and Jesser[11] have presented a unified theory which involves frictional forces, which contains the excess stress theory, and which gives good agreement with experiments on very-low-strained (<0.002) GaAsP films.

In this paper a frictional force is shown to account for the known properties of highly strained InGaAs on GaAs. These include a rapid transition from a small to an extremely large critical thickness over a small temperature range; an accompanying island-to-layer growth-mode transition; an equivalent growth-mode transition which occurs at constant temperature but with changing indium concentration; and strong evidence that island growth is nucleated by misfit dislocations.

In general there are serious difficulties in comparing critical-thickness and plasticity theories with experiment. The theories include processes described by exponential terms which may vary from insignificant to dominant over the experimental conditions of interest. The strategy here is to investigate regions of the available data which are so extreme in their behavior that they isolate only one factor; in this case a frictional force. Highly strained films ($\epsilon > 0.02$) have small critical thicknesses (~ 10 monolayers) and so dislocation velocity and multiplication effects are expected to be less significant than in thicker films with lower stresses. The data[12] are shown in Fig. 1, where the critical thicknesses of highly strained ($\epsilon = 0.023$) $In_{0.33}Ga_{0.67}As$ films are plotted against growth temperature. As the temperature is lowered there is a slow increase in critical thickness which then increases abruptly beyond the experimental resolution at 470 °C: No change in the surface lattice constant was detected. Also shown are data[13] for $x = 0.25$ with much thicker and lower-strained ($\epsilon = 0.018$) films. The data of

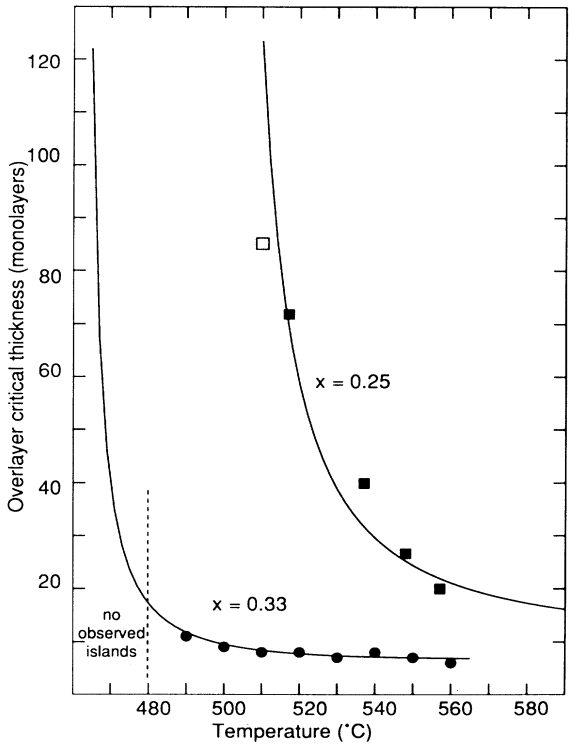

FIG. 1. Comparison of the h_c derived from Eq. (2) with experiment. The region of no island growth refers to the $x = 0.33$ data of Ref. 12 only. The $x = 0.25$ data (Ref. 13) were augmented by a point from an $x = 0.26$ experiment (Ref. 12) marked by the open square. The solid curves are given by Eq. (2) with $U = 1.0$ and 2.3 eV for $x = 0.25$ and 0.33, respectively.

Ref. 13 were augmented with one point from Ref. 12. Both sets were taken by reflection high-energy electron diffraction (RHEED). The $x=0.25$ data were found from measurements of the occurrence of island growth as shown by three-dimensional contributions to the diffraction pattern. Previous work[2,14] has shown that this growth-mode transition coincides with lattice expansion of the InGaAs as measured by RHEED. The $x=0.33$ data[12] were obtained by measurement of the surface lattice constant with a resolution of 0.003 Å. This equals a misfit resolution of 5×10^{-4} which is so small that for the present purposes it can be ignored.

Theory must account for a sharp transition in critical thickness over 20 °C. A simple force theory is known to approximate the critical thickness at 530 °C for a range of x. It has the form[7]

$$F_\epsilon - F_l \pm F_s = 0, \quad (1)$$

where $F_\epsilon = 21.0h\epsilon$, $F_l = 5.2[\ln(h/b)+1]$, and $F_s = \Gamma b \times \sin 60°$ are the forces in newtons on a misfit dislocation due to the elastic stress, the dislocation line tension, and the creation or loss of surface. The thickness of the overlayer is h and $b=4$ Å is the magnitude of the Burgers vector of a 60° misfit dislocation. The force F_s (Ref. 15) is included to obtain the correct value of the critical thickness at high temperature (~530 °C) and high strain with Γ, the surface energy, treated as an adjustable parameter. It is found[2] that $\Gamma \sim 1$ J m^{-2}, in agreement with experiment.[16] The constants are for $x=0.33$ but are only weakly x dependent. This equation is solved for the critical thickness h_c by putting $\epsilon = f$, where f is the misfit. At higher temperatures (> 560 °C) there is indium evaporation. At lower temperatures, the critical thickness is not given by (1), as it has no temperature dependence. It has previously been pointed out[6,7] that other forces should be included where appropriate, such as forces between gliding dislocations, dislocations and obstacles, and the force to move dislocations by glide. Fox and Jesser[11] have used terms for glide and atmosphere pinning. Here we will use a simplified version of the originally proposed[6] force for dislocation activation $F_f = Ah\exp(U/kT)$, where U is expected to be close to the Peierls energy and A is a constant. Thus Eq. (1) becomes

$$F_\epsilon - F_l + F_s - F_f = 0. \quad (2)$$

By choosing parameters A and U to fit the data at high temperatures and misfit we can investigate the lower-temperature transition region. Strain-versus-thickness curves derived from Eq. (2) are shown in Fig. 2. The curves have identical form shifted vertically with temperature. At high temperatures there is little variation in the critical thickness h_c, which is given by the intersection of the misfit f with the curves. As the temperature is lowered below 490 °C, the h_c rapidly increases until a temperature is reached where there is no intersection with f: The temperature is too low to activate dislocation movement. This is a major result of this theory: It predicts the rapid rise to an infinite critical thickness.

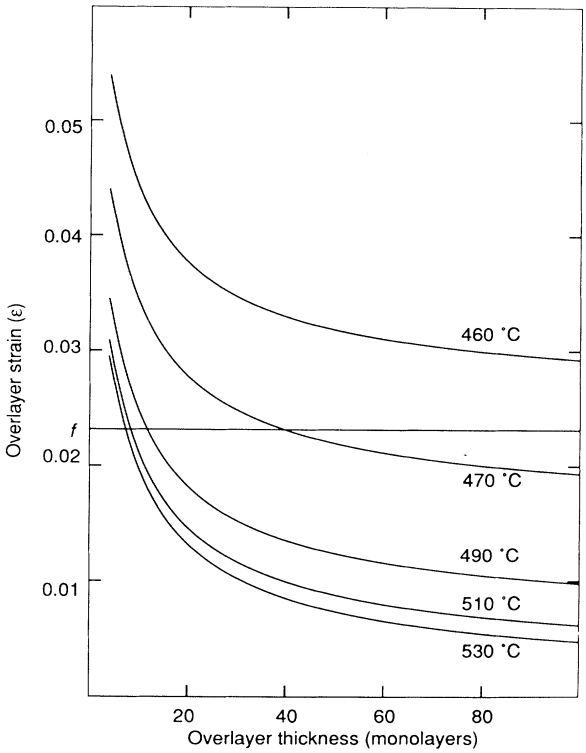

FIG. 2. Plots of strain against thickness for different temperatures predicted by Eq. (2) for an $In_{0.33}Ga_{0.67}As$ overlayer with U of 2.3 eV. The misfit on GaAs is marked by f.

The h_c derived from Eq. (2) are compared with experiment in Fig. 1. The $x=0.33$ curve was fitted to the data and a cutoff of 465 °C. A range of $U=2.0\pm0.5$ eV was found with the curve shown corresponding to $U=2.3$ eV. The data range is too small to obtain a precise value of A: $\ln A = -30\pm10$. The $x=0.25$ curve is the best fit to the points shown with $U=1.0$ eV: There are no experimental data on the cutoff temperature. Both energies are in the range of Peierls energies for semiconductors[17] and show that Eq. (2) gives a first-order description of the rise in critical thickness with temperature. The $x=0.33$ curve would be better fitted with a 5 °C higher cutoff temperature. More data are required before a choice of a more elaborate F_f needs be made: Fig. 2 shows that the cutoff region is highly sensitive to the exact position of f and the shape of the strain curve and so only a small change in the theory would be required. On the other hand the $x=0.25$ data set does not follow the functional form of the theory. These films are an order of magnitude thicker with lower strain and are out of the region where F_f can be isolated. The $x=0.25$ data do not provide support for the theory, but they are consistent with it.

Equation (2) predicts the rapid rise in critical thickness through a combination of a hyperbolic and exponential dependence on temperature. A temperature dependence which depends on exponential terms alone gives too high an activation energy. An example is the theory of Dodson and Tsao[8] in which the critical thickness depends on the time taken for an overlayer to relax. This time can be written[18] as

$$t(\delta,T)=Ce^{U/kT}\int_{\delta_0}^{\delta}\frac{d\delta'}{(\delta_{\rm equ}-\delta')^2(\delta'+\delta_0)}, \qquad (3)$$

where $\delta=f-\epsilon$ is a measure of the overlayer relaxation, $\delta=\delta_{\rm equ}$ when Eq. (1) is satisfied, C is a constant, U is a glide activation energy, and δ_0 is the initial relaxation of the overlayer due to grown-in dislocations. A lower bound on U can be estimated by comparing the times to relax on either side of the transition. Considering that the film growth rate is ~ 1 monolayer sec^{-1}, the measurement time is $\sim 10^2$ sec, and the total experiment time is $>10^3$ sec, a conservative lower bound for U can be found from $t(\delta_{\rm equ},743)>10^2 t(\frac{1}{2}\delta_{\rm equ},763)$, where the use of $\frac{1}{2}\delta_{\rm equ}$ avoids possible lengthy asymptotic approaches to $\delta_{\rm equ}$. This inequality gives $U>10$ eV if the ratios of the integrals (~ 5) are ignored and $U>15$ eV if they are included. These energies are too high for dislocation activation. The transition can be sharpened if $\delta_{\rm equ}$ is derived from Eq. (2) rather than from Eq. (1) for as f becomes tangential to $\epsilon(h)$, the term $(\delta_{\rm equ}-\delta)^2$ rapidly decreases. However, calculation shows that the activation energy still remains too high.

Equation (2) will now be applied to island growth. Of interest is the transition between layer and island growth modes for different misfits. It will be argued, on the basis of a simple growth model which describes the general experimental behavior of these strained overlayers, that while δ defines the state of relaxation of these films, $d\delta/dh$ is the parameter which determines the island growth transition. The problem then reduces to deriving this parameter as a function of x from Eq. (2). The experimental observations which must be fitted are the following: Three-dimensional growth is observed with RHEED within a monolayer of h_c.[2,12] Experiments over the range $0.3\leq x\leq 0.5$ show that these three-dimensional features attain a maximum within a few monolayers and then revert to layer growth.[2] If a thick layer is grown at a low temperature where islands do not form and the temperature is then raised, three-dimensional features appear.[12] The misfit value of ~ 0.02 is a boundary below which, even at high temperatures, three-dimensional features do not appear.[12] In what follows, a two-parameter model which fits these observations will be described and used to calculate the effect of strain on the island-to-layer transition. The assumptions[19] of the model are that (1) islands are nucleated by misfit dislocations, (2) each monolayer is grown in either the layer or island growth mode, and (3) dislocations which nucleate an island do not contribute to island nucleation later in the growth. Assumption (1) is supported by the simultaneous appearance of island growth and lattice relaxation. Detailed nucleation mechanisms will not be discussed here but it should be noted that dislocated islands within a strained layer can have a lower energy than a unformly dislocated overlayer[20,21] and the screw component of the 60° dislocations can overcome kinetic barriers to three-dimensional growth. Let λ be a surface diffusion length which characterizes the perturbation of the layer growth by the misfit dislocations and let the length ξ be dependent on the spacing between the misfit dislocations which are introduced by each monolayer: $\xi=\xi(1/(d\delta/dh))$. These parameters are defined so that for layer growth $\xi>\lambda$ and for island growth $\xi\leq\lambda$. (This is analogous to heterogeneous thin-film growth[22] with λ the catchment radius of strongly trapping heterogeneities.) Below h_c, $d\delta/dh=0$ and $\xi\gg\lambda$; hence there is layer growth. At h_c, $d\delta/dh$ is a maximum (see Fig. 2) and then decreases approaching zero as h increases. If $\xi<\lambda$ at h_c, islands will form; but as the growth proceeds ξ will become greater than λ and layer growth will resume as observed. The distance between dislocations ($b/2\delta$ for 60° dislocations) is plotted in Fig. 3 as a func-

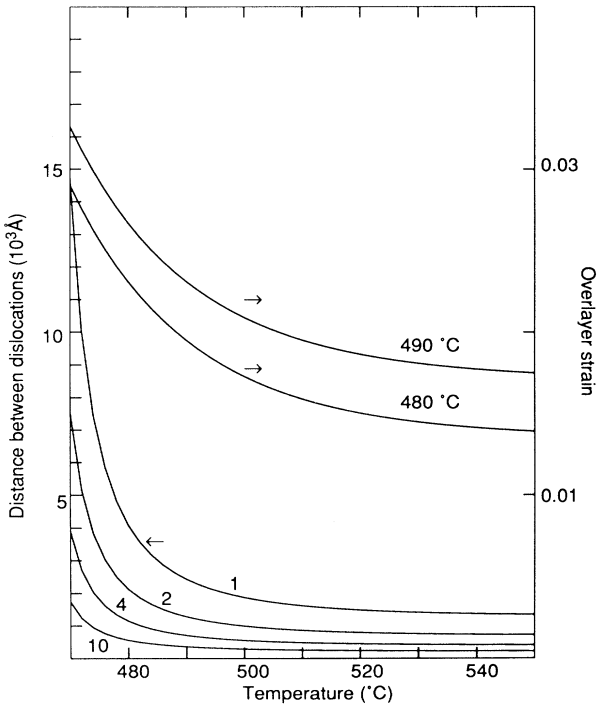

FIG. 3. Left ordinate: distance between misfit dislocations plotted against temperature after 1, 2, 4, and 10 monolayers of $In_{0.33}Ga_{0.67}As$ have been deposited in addition to h_c on GaAs. Right ordinate: curves of misfit strain against temperature with constant values of $d\delta/dh$ equal to those at h_c for $x=0.33$ and for the temperatures indicated.

tion of temperature for fixed numbers of monolayers past h_c using Eq. (2). The increase in dislocation spacing occurs as f becomes tangential to $\epsilon(h)$. As the temperature is lowered, $d\delta/dh|_{h_c+}$ decreases rapidly until $\lambda < \xi|_{h_c+}$ and layer growth occurs. This accounts for the no-island region shown in Fig. 1. The value of λ is unknown; estimates of diffusion lengths on III-V semiconductors range from a few hundred angstroms (~ 4 Å$\exp[(0.3\text{ eV})/kT]$),[23] for two-dimensional nucleation in the presence of vicinal steps, to microns[24] for linear diffusion. The former is more likely to fit the present circumstances and has the right magnitude. The assumption of constant λ in this model is a reasonable approximation because of the much stronger temperature dependence of $d\delta/dh|_{h_c+}$.

This model is now applied to the variation of the island-to-layer growth transition with x or misfit. If we ignore the weak dependence of the constants in Eq. (2), then only the misfit f is a function of x and the curves of Fig. 2 are not only of identical form but are identical to the $\epsilon(h)$ for all x. Consequently the intersection of a line of constant h with the curves of Fig. 2 has a constant $d\delta/dh|_{h_c+}$ for all T, with the coordinates of the intersection $(h, f(x))$. A plot of the misfit strain against temperature for two $d\delta/dh$ characteristic of the edge of the $x=0.33$ transition of Fig. 1 is given in Fig. 3. For misfits below these curves, there will not be island growth, since $\xi > \lambda$ at the critical thickness and can only increase with h. (This can best be seen by dropping a vertical line from the intersection of f and the 490 °C curve of Fig. 2.) Figure 3 shows that this cutoff of island growth against strain occurs below $f=0.02$, in agreement with experiment. Thus, in conclusion, the incorporation of temperature dependence through F_f and the identification of $d\delta/dh$ as the characterizing parameter for island growth have given agreement between theory and experiment for both critical-thickness and growth-mode transitions in highly strained InGaAs.

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