Dislocation dynamics in strain relaxation in GaAsSb/GaAs heteroepitaxy

B. Pérez Rodriguez
Randall Laboratory, Applied Physics Program, University of Michigan, Ann Arbor, Michigan 48109

J. Mirecki Millunchick
Department of Materials Science and Engineering, University of Michigan, Ann Arbor, Michigan 48109-2136

(Received 11 January 2006; accepted 27 May 2006; published online 18 August 2006)

The real-time stress evolution has been investigated during molecular-beam epitaxial growth of GaAs$_{1-x}$Sb$_x$/GaAs metamorphic buffer. These real-time data were obtained using an in situ multibeam optical sensor measurement and has been combined with detailed analysis of data obtained from x-ray diffraction, transmission electron microscopy, and atomic force microscopy. We compare the strain relaxation of two different compositions of GaAs$_{1-x}$Sb$_x$, and correlated the development of dislocation structure and morphology. Several distinct stages of the strain relaxation were observed during growth, which can be separated in three main regimes: pseudomorphic growth, fast strain relaxation, and saturation. Transmission electron microscopy data show that GaAs$_{0.61}$Sb$_{0.39}$ buffer layers have a larger fraction of pure-edge dislocations that arise during the earliest stages of growth. This could have a significant influence in the fabrication of buffer layers, since pure edges are favored over the threading dislocations. The strain relaxation profile for each film was modeled using a modified model of Dodson and Tsao [Phys. Rev. B 38, 12383 (1988)] that takes into account the elastic interactions of misfit dislocations. The model results agree with the experimental data and show that interaction of misfit dislocations is responsible for the large residual stress. In addition, following the description developed by Dodson and Tsao [Phys. Rev. B 38, 12383 (1988)] for the rate of dislocation multiplication, we were able to determine the line density of threading dislocations from the experimental data. This has a potential application in the design of metamorphic buffer layers because our observations are made in real time on individual growth, without the need of external characterization to measure the dislocation density. © 2006 American Institute of Physics. [DOI: 10.1063/1.2245206]

I. INTRODUCTION

Metamorphic buffer layers have recently been proven to be a useful way to integrate high performance devices with low cost substrates. The success of the metamorphic approach is in the ability of the buffer layer to relax the stress due to lattice mismatch between the substrate and the epitaxial layer, without degrading the quality of the active region. Stresses that build up in the buffer layer as the film grows have been shown to result in bending of the substrate, which has been studied for SiGe/Si, Ge/Si, Fe/GaAs, InAs/GaAs, and InGaAs/GaAs (Ref. 6) films. There are several mechanisms by which the stress (or the strain) may be relieved. It is well known that the introduction of dislocations relaxes the strain, however, the presence of these defects in the active region degrades device properties. Therefore, one requirement for an appropriate metamorphic buffer layer is that the defect density be minimized near the active region. Also, the surface has been observed to roughen in response to the stress or in the presence of buried dislocations. The result of such roughening may be in higher carrier scattering, resulting in reduced mobility. Consequently, understanding and controlling the origin of stress is of importance to the reliable fabrication of devices upon metamorphic buffer layers. In this work, the stress evolution and strain relaxation for GaAs$_{0.5}$Sb$_{0.5}$ and GaAs$_{0.61}$Sb$_{0.39}$ films deposited on GaAs (001) substrates is examined and correlated to evolution of the surface morphology and the formation of misfit dislocations. The stress evolution is also compared to a modified Dodson-Tsao model that takes threading dislocation interactions into account. This model is shown to predict the linear density of dislocations within a factor of 2 or 3.

II. EXPERIMENT

GaAs$_{1-x}$Sb$_x$ films were grown at a substrate temperature $T_{sub}=477$ °C using molecular-beam epitaxy (MBE) on 3 in. Si-doped GaAs (001) substrates with a thickness of 450 μm. Whole wafers were loaded on non-In-bonded sample holders to allow flexure during growth. The oxide was desorbed under an As$_2$ overpressure at $T_{sub}=600$ °C, followed by the deposition of a 1470-Å-thick GaAs buffer layer at $T_{sub}=573$ °C. Incorporation rates $R$ were calibrated using reflection high-energy electron diffraction (RHEED) prior to the growth of the film. The incorporation rates used throughout the experiment for Ga and As were $R_{Ga}=0.96$ and $R_{As}=1.11$ monolayer/s (ML/s), respectively. These high rates were used to avoid any compositional variations across the film. The Sb incorporation rate for the GaAs$_{0.5}$Sb$_{0.5}$ film was $R_{Sb}=0.61$ ML/s, and $R_{Sb}=0.41$ ML/s for the GaAs$_{0.61}$Sb$_{0.39}$ film. The stress in the GaAs$_{1-x}$Sb$_x$ film was

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4 Electronic mail: joannamm@umich.edu
further characterized using transmission electron microscopy. The microstructure, including the dislocation density, was characterized in situ by measuring the wafer curvature during growth as described in Refs. 11 and 12. The stress-thickness evolution during the growth of the GaAs$_{0.5}$Sb$_{0.5}$/GaAs film is shown in Fig. 1(b). This film is in contrast to reports in other systems such as In$_{0.2}$Ga$_{0.8}$As and In$_{0.12}$Ga$_{0.88}$As$_{0.99}$N$_{0.01}$, where the apparently large fluctuations in the strain relaxation for $h_f<25$ Å are due to the fact that the strain is calculated from the product of the stress thickness and the lattice parameter of the substrate and film, respectively. The thermally induced strain may be obtained from x-ray diffraction (XRD) and MOS data, which is given by:

$$\gamma = \left(\frac{f_o - f_s}{f_o}\right) \times 100, \quad \text{where} \quad f_o = (a_s - a_f)/a_s \quad \text{is the misfit strain, and} \quad a_s \quad \text{and} \quad a_f \quad \text{are the lattice parameters of the substrate and film, respectively.}$$

An additional relaxation may occur during cooling. Figure 2 shows the stress evolution versus time during cooling from 470 °C to 200 °C. The inset shows the first 200 Å of the growth, and the straight line corresponds to a coherently strained film at this composition, and it is evident that for $h_f<25$ Å no relaxation takes place. This linear increase corresponds to the pseudomorphic regime. As the film gets thicker the slope appreciably decreases due to strain relaxation. The evolution of the strain relaxation during growth is shown in Fig. 1(b), and is defined as $\gamma = (f_o - e / f_o) \times 100$, where $f_o = (a_s - a_f)/a_s$ is the misfit strain, and $a_s$ and $a_f$ are the lattice parameters of the substrate and film, respectively. The thermally induced strain may be obtained from x-ray diffraction (XRD) and MOS data, which is given by:

$$\gamma = \frac{\Delta \delta}{\delta} \times 100,$$

where $\alpha_S = 6.93 \times 10^{-6}$ K$^{-1}$, the coefficient of thermal expansion of the GaAs substrate, $\alpha_F = 7.90 \times 10^{-6}$ K$^{-1}$, the coeffi-
cient of thermal expansion of the GaAs$_{0.5}$Sb$_{0.5}$, interpolated from the values of pure GaAs and GaSb, and $\Delta T =-270$ K, the drop in the temperature during cooling. Since the substrate is much thicker than the film, we assume that the difference in temperature between substrate and film is negligible. The induced strain thus calculated is $\epsilon_{th}=0.26 \times 10^{-3}$, which corresponds to an induced tensile stress $\sigma_{th}$ =0.03 GPa, and is in reasonable agreement with the measured stress. The remainder of the stress in the film is likely generated during cooling from $T=200$ °C to room temperature. The predicted induced stress for In$_{0.2}$Ga$_{0.8}$As and In$_{0.12}$Ga$_{0.88}$As$_{0.99}$N$_{0.01}$ at this cooling rate is $\epsilon_{th}=0.01$ GPa and $\sigma_{th}=-0.005$ GPa, respectively. In both cases, the magnitude of the thermal stress is quite small, and the sign of the stress is negative. Therefore, further relaxation in these films is not expected upon cooling. Thus, the MOS and XRD data for those materials systems should be nominally the same, in agreement with the published experimental data. The morphology at various points of the strain evolution was also examined. Figure 3 shows AFM micrographs of films at various points along the strain relaxation curve. These data show that the surface topography develops quite differently within each of these three strain relaxation regimes. The boundary between the pseudomorphic and the fast relaxation regime [Fig. 3(a)] is characterized by the nucleation of large island of 0.5–1.0 μm long by 0.2 μm wide elongated along [110]. These large islands are not likely oval defects because the appearance of these features depends on group V flux and their density decreases as the film thickness increases, while the density of oval defects has been observed to be independent of the III/V flux ratio and to increase with film thickness. Furthermore, oval defects vary in size between 3–15 μm, contrary to what we observe. During the fast relaxation regime, smaller isotropic island of approximately 82 nm in diameter nucleates, as seen in Fig. 3(b). Figure 3(c) shows a decrease in the smaller island density due to coalescence as the strain relaxation reaches saturation. The surface remains rough at large thickness [Fig. 3(d)] likely due to the enduring coherent strain.

We have examined the dislocation structure at the epilayer/substrate interface using (110) cross-sectional high resolution TEM (HRTEM). Figure 4 shows a Fourier-filtered (110) cross-sectional HRTEM image for GaAs$_{0.5}$Sb$_{0.5}$ film at a thickness $h_f=65$ Å. Dislocations in this image are identified by arrows along the {111} and {110} planes. Two pure-edge dislocations, which are distinguish by an extra {110} plane, and three 60° dislocations, which have one extra {111} half plane, are apparent in this image. The dislocation density was determined using this Fourier-filtered technique for several sections taken from the film near the beginning of the fast relaxation regime ($h_f=65$ Å), and another at the end ($h_f=342$ Å). The line density of each type of dislocation and average spacing $p$ are summarized in Table I. For the $h_f=65$ Å film, the linear density of 60° dislocations is higher than the density of pure-edge dislocations by nearly a factor of 2. For the $h_f=342$ Å thick film, the density of 60° dislocations doubles, while the density of pure-edge dislocations quadruples, indicating that pure-edge dislocations have a higher nucleation rate in the fast relaxation regime. This re-

TABLE I. Summary of the density of different kinds of dislocations in GaAs$_{1-x}$Sb$_x$ observed by HRTEM.

<table>
<thead>
<tr>
<th>% of Sb content</th>
<th>Thickness (Å)</th>
<th>60°</th>
<th>Pure edge</th>
<th>$p$ (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>65</td>
<td>1.3</td>
<td>0.7</td>
<td>83±13</td>
</tr>
<tr>
<td>50</td>
<td>342</td>
<td>2.7</td>
<td>2.6</td>
<td>70±5</td>
</tr>
<tr>
<td>39</td>
<td>1818</td>
<td>1.7</td>
<td>1.2</td>
<td>100±14</td>
</tr>
</tbody>
</table>
The average dislocation line density for the threading dislocations for various thicknesses. The average dislocation line density was found to be $1.1 \times 10^5 \text{ cm}^{-2}$ at $h_f=2908 \text{ Å}$ over a range of approximately 6.8 µm. For thicknesses beyond $h_f=2908 \text{ Å}$, TEM images do not reveal a significant increase in the threading dislocation line density. The areal dislocation density using plan-view TEM are $7.9 \times 10^5 \text{ cm}^{-2}$ for $h_f=2908 \text{ Å}$ and $1 \times 10^{10} \text{ cm}^{-2}$ for $h_f=7100 \text{ Å}$. This suggests that dislocation interactions dominate once this high linear density is achieved. Several mechanisms are possible that can prevent complete strain relaxation, such as annihilation between edge dislocations or repulsive interactions between dislocations.

The stress evolution for lower Sb composition layers was also examined. We grew GaAs$_{0.61}$Sb$_{0.39}$/GaAs layers, which have a lattice mismatch of $f_o=3.1\%$, to a thickness of $h_f=1818 \text{ Å}$. The lower Sb composition was obtained by changing the Sb valve position until an incorporation rate of $R_{\text{Sb}}=0.41 \text{ ML/s}$ was reached, while keeping the Ga, As, and growth temperature at the same values as for the GaAs$_{0.61}$Sb$_{0.39}$ layer. Figure 6 shows a comparison of the strain relaxation for the GaAs$_{0.61}$Sb$_{0.39}$ (solid line) and GaAs$_{0.5}$Sb$_{0.5}$ (dotted line) films. As for the more highly mismatched film, the strain relaxation evolution can be divided into three regimes: pseudomorphic, fast relaxation, and saturation. For the GaAs$_{0.61}$Sb$_{0.39}$ film, the pseudomorphic regime is larger, $h_f<160 \text{ Å}$, as expected due to its lower lattice mismatch. Fast relaxation occurs for $160 \text{ Å}<h_f<413 \text{ Å}$, where the film relaxes up to 47% of the strain with a rate of 0.2% /Å. Beyond 413 Å, the rate of strain relief continues to decay for the duration of the growth to approximately 0.015% /Å. At $h_f=1725 \text{ Å}$, roughly 82% of the strain has relaxed. Interestingly, the lower mismatch film relaxes at a faster rate in the saturation regime, resulting in a lower residual strain than its higher misfit counterpart.

The morphology at various thicknesses was examined for the GaAs$_{0.61}$Sb$_{0.39}$ films. Figure 7(a) shows the nucleation of large islands upon a smooth wetting layer, similar to the morphology observed for GaAs$_{0.5}$Sb$_{0.5}$ in the pseudomorphic regime [Fig. 3(a)], except that the density of these islands is lower. As the thickness of the film increases, the islands form pits at their center, and small islands with an average diameter of 62 nm form, as seen in Fig. 7(b). During the saturation regime, the smaller islands coalesce forming ridges along [110], and the large islands disappear [Fig. 7(c)]. Cross-sectional TEM images over a range of 2.7 µm indicate a lower density of dislocation walls, thus a lower density of pure-edge dislocations, as seen in Fig. 8. From these images, the line density of threading dislocations was found to be $2.2 \times 10^5 \text{ cm}^{-1}$. An areal threading dislocation density of $5.39 \times 10^6 \text{ cm}^{-2}$ was measured using plan-view TEM for a
GaAs$_{0.61}$Sb$_{0.39}$ film at $h_f = 1818$ Å. Table I shows a smaller density of pure-edge dislocations relative to the obtained for GaAs$_{0.5}$Sb$_{0.5}$ films, while the number of threading dislocations are on the same order of magnitude.

**IV. DISCUSSION**

The dynamics of dislocation motion are crucial in the strain relaxation in thin films. It has been postulated that dislocations move through the film by a combined effect of glide and climb.$^{23-26}$ Dodson and Tsao have shown that the strain relaxation rate may be expressed as a single nonlinear differential equation in thickness $h$,

$$\frac{d\gamma}{dh} = \frac{CM^2}{r_g \sigma_o} \left[ f_o - \gamma(h) - r(h) \right] \left[ \gamma(h) + \gamma_o \right],$$

where $r_g$ is the growth rate, $f_o$ is the lattice mismatch, and $\gamma_o$ is a fitting parameter that represents the initial strain relief due to preexisting misfit dislocations. $CM^2/\sigma_o^2$ is a fitting parameter where $C \approx e^{-U/kT}$. The term $r(h)$ is the equilibrium residual strain arising due to the dislocations. Because pure-edge dislocations are sessile, we only consider the contribution of the mobile threading dislocations. Using the classic treatment from Hirth and Lothe, the equilibrium residual strain may be calculated,$^{27}$

$$r(h) = \frac{b_{60} (1 - \nu \cos^2 \theta)}{8\pi(1 + \nu)R_{h,p} \cos \lambda} \ln \left( \frac{\alpha_r R_{h,p}}{b} \right),$$

where $b_{60} = a/2\sqrt{2}$ is the Burger’s vectors of threading dislocations, $\nu$ is the Poisson ratio, $\alpha_r$ is the core cutoff parameter, and $\theta$ is the angle between the dislocation line and the Burger’s vector. $R_{h,p}$ is called the complex modulus of dislocation semispaning, which is the approximate solution for a stress-free boundary surface associated with two free cylinder surfaces. When we consider only the image dislocation outside the film, the self-stress of a misfit dislocation is $R_{h,p} - h$, where $h$ is the film thickness. However, as Fischer et al. point out,$^{28}$ dislocations in the film not only interact with their image, but with the real stress fields of misfit dislocation in the interface. For an ideal case, the complex dislocation semispaning term for the self-stress of a real symmetrical interfacial misfit dislocation is approximately one half of the dislocation spacing.$^{27}$ However, due to the uneven distribution of the stress field and the asymmetry of the dislocation spacing in the interface, the complex dislocation semis-
pace term for a real misfit dislocation may be approximated \( R_{b,p} \sim p \), where \( p \) is the misfit dislocation spacing. Taking the interactions between the image dislocations and the dislocations in the interface,28 the complex modulus becomes

\[
R_{b,p} = \left( \frac{1}{h^2} + \frac{1}{p^2} \right)^{1/2}.
\]

It should be noted that in this form of the equation \( R_{b,p} \sim h \) for small thicknesses, and \( R_{b,p} \sim p \) for large thicknesses. The \( p \) may be measured experimentally by TEM or XRD for each sample. However, we have found that these values compare well to the predicted value of \( p \) based solely upon lattice mismatch considerations as described by the expression

\[
p = \frac{a_{\text{Film}}a_{\text{Sub}}}{2(a_{\text{Film}} - a_{\text{Sub}})}.
\]

The results of this model with and without the dislocation interaction are plotted in Fig. 9(a) and compared to the strain relaxation data for GaAs\(_{0.5}\)Sb\(_{0.5}\). As shown in previous reports,11,29 the model does not agree well with actual strain relaxation data beyond the fast relaxation regime when \( R_{b,p} \) is taken to be the thickness of the film. However, when \( R_{b,p} \) takes dislocation interactions within the interface into account, the model fits the data well into the saturation regime. The fidelity of the fit degrades by 4%–6%, however, at the largest thicknesses studied here.

It is now possible to obtain linear dislocation densities from these fits. The line density of misfit dislocation is related to the strain relief \( \gamma \) by

\[
\gamma = b\rho \cos(\lambda).
\]

Using the phenomenological description of the rate of dislocation multiplication described by Dodson and Tsao,26 the linear dislocation density is given by

\[
\rho = \frac{d\gamma}{dt} = \frac{1}{b\cos(\lambda)}\left(CM^2/\sigma_o^2\right)e_{\text{eff}}^2,
\]

where the effective stress \( e_{\text{eff}} \) and strain relief rate \( d\gamma/dt \) are the \textit{in situ} strain measurements and \( b \cos(\lambda) = b_{60} \). Table II compares the dislocation line densities obtained experimentally from TEM images to those predicted by the model. In the saturation regime of GaAs\(_{0.5}\)Sb\(_{0.5}\) film, the estimated dislocation line density \( \rho_{\text{model}} = 7.4 \times 10^4 \) cm\(^{-1}\), which is in reasonable agreement with the experimental values \( \rho_{\text{exp}} = 1.1 \times 10^5 \) cm\(^{-1}\). Similarly, for GaAs\(_{0.61}\)Sb\(_{0.39}\), the dislocation line density was estimated to be \( \rho_{\text{model}} = 1.9 \times 10^5 \) cm\(^{-1}\) which is also in very good agreement with the experimental values of \( \rho_{\text{exp}} = 2.2 \times 10^5 \) cm\(^{-1}\). In order to verify that the model is applicable to other material systems, we also fit the experimental data for the strain relaxation for In\(_{0.2}\)Ga\(_{0.8}\)As grown on GaAs. In all the three cases, the experimentally observed values are about a factor of 2 higher than the predicted values. This is consistent with the observation that the models consistently underestimate the strain relaxation in these films. Another source of the discrepancy may be in the experimental measurement technique. The strain relaxation data from which the dislocation density is predicted are taken over a large portion of the sample (typically a few square centimeters), whereas the dislocation density is measured from several TEM samples and represent a much smaller area of the sample. Nonetheless, this model shows that the linear dislocation density can be estimated in real time, without the necessity of etching or sample preparation for TEM imaging.

V. CONCLUSIONS

The real-time strain relaxation was investigated in GaAs\(_{1-x}\)Sb\(_{x}\) films with nominal Sb compositions of \( x = 0.5 \)

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|c|}
\hline
Film & \( d\gamma/dt \) (s\(^{-1}\)) & \( e_{\text{eff}} \) & CM\(^2/\sigma_o^2 \) (s\(^{-1}\)) & \( \rho_{\text{model}} \) (cm\(^{-1}\)) & \( \rho_{\text{exp}} \) (cm\(^{-1}\)) \\
\hline
GaAs\(_{0.5}\)Sb\(_{0.5}\) & 7.61 \times 10^{-7} & 4.06 \times 10^{-3} & 30 & 7.4 \times 10^4 & 1.1 \pm 0.3 \times 10^5 \\
GaAs\(_{0.61}\)Sb\(_{0.39}\) & 7.54 \times 10^{-6} & 5.53 \times 10^{-3} & 62 & 1.9 \times 10^5 & 2.2 \pm 0.6 \times 10^5 \\
In\(_{0.2}\)Ga\(_{0.8}\)As & 3.98 \times 10^{-7} & 2.54 \times 10^{-3} & 110 & 2.8 \times 10^4 & 4.9 \pm 2.5 \times 10^4 \\
\hline
\end{tabular}
\caption{Comparison of the threading dislocation line density obtained experimentally and from the model.}
\end{table}
and $x=0.39$. The strain relaxation goes through several regimes, which are correlated to the surface morphology and the nucleation of dislocations. The fast relaxation regime is characterized by the nucleation of small islands and an abrupt increase in the number of edge dislocations, while the saturation regime is distinguished by coalescence of small islands. The data were compared to a dislocation kinetic model developed by Dodson and Tsao with the addition of a dislocation interaction term, which results in a reasonable fit to the data. Using this model, estimated line dislocation densities are in good agreement with the values obtained experimentally. These results indicate that it is possible to estimate the dislocation density in real time using this wafer curvature method.

ACKNOWLEDGMENT

The work reported here is supported by a grant from the National Science Foundation under Award No. DMR0092602.