

**X-ray studies of Si/Ge/Si(001) epitaxial growth with Te as a surfactant**

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(Received 13 June 2002; published 14 January 2003)

X-ray standing waves (XSW) and grazing incidence x-ray diffraction (GIXD) were used to investigate the crystallinity of ultrathin Ge films grown by molecular-beam epitaxy on Si(001) with and without Te as a surfactant. The Ge layer thickness ranged from 1 to 10 ML. The results clearly indicate that Ge films grown with Te have a higher degree of crystallinity compared to those grown without Te. For example, GIXD shows that 9 ML Ge grown on Si(001) with Te is strained in plane; while the same film grown without Te is relaxed. The (004), (022), and (008) XSW results are used to determine the registry of the Ge atoms with respect to the Si lattice. This is compared with macroscopic continuum elasticity theory predictions for Ge/Si(001).

DOI: 10.1103/PhysRevB.67.035404

PACS number(s): 68.55.Ac

**INTRODUCTION**

Si/Ge heterostructures continue to develop a greater presence in high-frequency field-effect transistors.<sup>1</sup> Therefore, the ability to grow thicker and higher-quality epitaxial Ge films on Si(001) continues to be a motivation for engineers and scientists. Surfactant mediated epitaxy (SME) has proven to be effective for improving Ge epitaxial quality by increasing the critical thickness of Ge grown on Si and thus, allowing the fabrication of abrupt interfaces in Si/Ge/Si heterostructures. Surfactants act to improve epitaxial growth by decreasing the adatom (in our case Ge) surface mobility and, thereby, promote two-dimensional (2D) epitaxial growth. A prerequisite for a surfactant in this case is to lower the surface free energy, thus, providing a driving force for the surfactant atom to site exchange with Ge adatoms as Ge is deposited. In addition to this, it is advantageous for the surfactant to have a low solubility in both Si and Ge. Most SME Ge/Si work has focused on group V elements [As,<sup>2-4</sup> Sb,<sup>5-7</sup> and Bi (Refs. 8, 9)]. However, group IV (Refs. 10, 11) and VI (Refs. 12, 13) elements have also been investigated. In this work, we have chosen Te to be used as the surfactant and have studied molecular-beam epitaxy grown Ge films between 1 and 10 ML grown with and without a surfactant.

The growth of Ge on Si(001) proceeds layer-by-layer for the initial 2 to 3 ML. Subsequent growth results in growth front roughening or islanding as a means of relieving the strain set up by the 4% lattice mismatch between Si and Ge.<sup>5,14</sup> By incorporating a surfactant in the growth process, one is able to grow coherently strained films well beyond 3 ML. These films have been reported to grow defect free up to ~10 ML, whereupon V-shaped defects consisting of (111) planes tilted perpendicular to the direction of maximum strain begin to form.<sup>4,12</sup> Supposedly, these defects relieve the misfit progressively and thus relieve strain in the film as it is grown. After ~50 ML, dislocations appear in the structure originating at the V-shaped defect sites. Higuchi *et al.* studied Te mediated Ge/Si(001) epitaxy with reflection high-energy electron diffraction (RHEED) and transmission electron microscopy (TEM).<sup>12,13</sup> They report that 550 Å thick Ge films grown with Te exhibit RHEED patterns that consist of well-developed streaks. This indicates 2D layer-by-layer growth and thus the suppression of islanding. Additionally,

cross-sectional TEM (Ref. 12) of these same samples shows the presence of stacking faults and two-phase diffraction patterns (Si and Ge) which suggest a variation of lattice parameter throughout the Ge film.

Our studies have focused on the initial growth (<20 Å) of Ge on Si(001). By using high-resolution x-ray standing waves (XSW's) we are able to determine the registry of Ge adatoms with respect to the Si(001) bulk substrate lattice and conclude that Te as a surfactant acts to substantially decrease the amount of disorder in Ge epitaxial layers. The x-ray standing wave technique has proven to be successful in characterizing and demonstrating that delta layers of Ge grown on Si with Sb as a surfactant are superior to non-SME samples.<sup>15,16</sup>

Grazing incidence x-ray diffraction (GIXD) is a complementary technique that can be used to map the strain distribution in thin films. Williams *et al.*<sup>17</sup> verified that the critical thickness for strain relaxation is between 3 and 4 ML for Ge growth on Si at 500 °C. For 10 ML thick films they found that the strain in the Ge epilayer consisted of both a fully relaxed component as well as a Si-Ge alloy component. Thornton *et al.*<sup>18</sup> performed a similar study using Sb as a surfactant and measured the critical thickness for the onset of Ge relaxation to be ~11 ML and claim that even up to 55 ML the films grown with Sb are at least partially strained. It is apparent from our data that the growth mode for the 9 ML sample grown with Te is pseudomorphic. This is in contrast to the sample with the same Ge coverage grown without surfactant, which is almost fully relaxed.

The Te/Si(001) 1×1 surface was used as the template for Ge deposition. Te, a hexavalent group VI element, can saturate all available surface dangling bonds of the group IV Si or Ge surface. This restores a passivated 1×1 terminated structure rendering it suitable for surfactant behavior. The Te/Si(001) structures consist of a saturation coverage (~0.8–1 ML) of Te atoms residing on bridge sites.<sup>19</sup> The ~0.8 ML Te/Ge(001) system has a similar structure.<sup>20,21</sup> Te rows are occasionally missing in both of these systems in order to accommodate the surface stress induced from the adsorption of the larger Te atom. The tendency for Te to passivate the Si and Ge(001) surfaces provides a driving force for Te to migrate to the growth surface during Ge/Si(001) heteroepitaxy. This is also confirmed from consider-

ation of binary phase diagrams,<sup>22</sup> where the binding energy is estimated to be stronger in Si-Ge or Ge-Ge than in Si-Te or Ge-Te. In contrast to other surfactants such as arsenic, tellurium's solubility in both Si and Ge is very low, thus, reducing the likelihood of any background doping in Si or Ge. Additionally, Te completely desorbs from Ge(001) at a moderate temperature (450 °C), therefore, making it relatively easy to remove the surfactant after growth.

### EXPERIMENT

The set of Si/Ge/Si(001) heterolayer structures were grown by molecular-beam epitaxy in a UHV system with a base pressure lower than  $1 \times 10^{-10}$  Torr. The Si substrates, which were 10 mm  $\times$  10 mm in area and 3 mm thick, were cleaned by the Shiraki method<sup>23</sup> and outgassed for at least 12 h at 650 °C. The samples were then annealed several times via indirect heating to 850 °C until a sharp two-domain  $2 \times 1$  low-energy electron diffraction (LEED) pattern was observed. Sample cleanliness was confirmed by Auger electron spectroscopy (AES) to ensure that the carbon and oxygen contamination was less than 0.01 ML. For the samples with Te, Te was deposited first onto a substrate held at 300 °C and then annealed to 400 °C for 10 min until a  $1 \times 1$  LEED pattern formed. Additionally, a Te overpressure of  $10^{-6}$  Torr was maintained during both Ge and Si growth in order to compensate for Te desorption. Ge was evaporated from a Knudsen cell at a rate of 0.1 ML/min with the substrate held at 410 °C. Continuing at 410 °C, the Si cap was deposited from an *e*-beam evaporator operating at 110 W with a corresponding growth rate of 1 ML/min. LEED and AES measurements were made after each step in the growth process. The Ge coverage of each sample, which ranged from 1 to 10 ML was verified by comparing its fluorescence signal to that of an ion-implanted standard. The Si cap thickness and interface roughness were determined by low-angle x-ray reflectivity.

The (004) and (022) XSW scans were performed at the NSLS X15A beamline. The measurements were made by monitoring the Ge  $K\alpha$  fluorescence signal while scanning in energy through the Si(004) and Si(022) rocking curves. The standing-wave field is generated by dynamical Bragg diffraction from the Si single crystal substrate. As the incidence angle (or energy) is scanned through the range of the Bragg reflection, the standing-wave nodal planes move inward one-half a  $d_{hkl}$  spacing. The coherent fraction and coherent position of the Ge atoms in the film is determined by applying dynamical diffraction theory analysis to the Ge  $K\alpha$  fluorescence yield data. A more detailed review of the XSW technique is available elsewhere.<sup>24</sup> The (008) XSW measurements were made at the DND-CAT 51D-C beamline of the Advanced Photon Source.

### RESULTS AND DISCUSSION

The structural parameters determined in our XSW measurement are coherent fraction ( $f_H$ ) and coherent position ( $P_H$ ) for Ge. These are, respectively, the amplitude and phase of the  $\mathbf{H} = hkl$  Fourier component ( $\mathcal{F}_H$ ) of the normal-

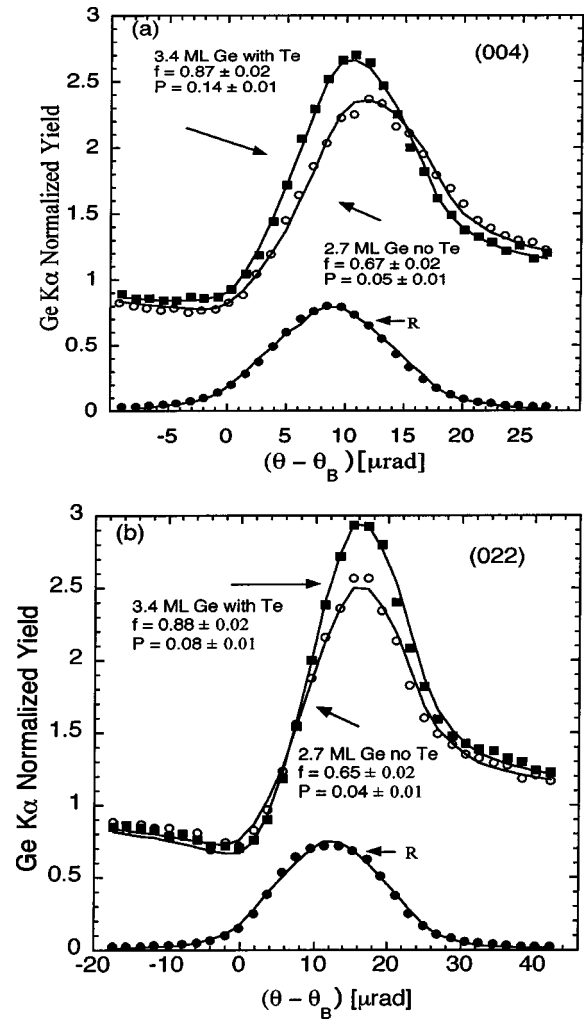


FIG. 1. The XSW experimental and theoretical angular dependency of the Ge  $K\alpha$  fluorescence yield and reflectivity collected while scanning through the (a) Si(004) and (b) Si(022) Bragg reflection. The Si/Ge/Si(001) samples are a 3.4 ML Ge film grown with Te as a surfactant compared with a sample that is a 2.65 ML Ge film grown with no surfactant.  $E_\gamma = 13.5$  keV.

ized Ge atomic distribution function,  $\rho(r)$ ,

$$\mathcal{F}_H = \int \rho(\mathbf{r}) e^{2\pi i \mathbf{H} \cdot \mathbf{r}} d\mathbf{r} = f_H e^{2\pi i P_H}. \quad (1)$$

The (004) XSW measurement probes the registry of the Ge atoms with respect to the Si substrate lattice along the growth direction while the (022) measurement probes a component that is sensitive to the registry both in and out of plane. Figures 1(a) and 1(b) compare fluorescence fits for two of the samples, one with and one without surfactant. The stronger modulation (higher coherent fraction) in Ge fluorescence for the sample grown with Te signifies a higher degree of ordering in the epitaxy.

The bulk lattice constant of Ge is 4.2% larger than that of Si at room temperature. Therefore, if the epitaxial layer is compressively strained to lattice match the Si substrate, the Ge film will be tetragonally distorted out of plane. Based on the elasticity theory, the strain in the [001] direction is given by,

$$\varepsilon_{\perp} = -2 \frac{c_{12}}{c_{11}} \varepsilon_{\parallel}, \quad (2)$$

where the elastic constants for Ge are  $c_{11}=12.4 \times 10^{10}$  N/m<sup>2</sup> and  $c_{12}=4.13 \times 10^{10}$  N/m<sup>2</sup>.<sup>25</sup> Therefore,  $\varepsilon_{\parallel} = -0.040$  results in  $\varepsilon_{\perp} = 0.027$ . This results in an out-of-plane Ge atomic layer spacing that is  $\delta_{004}^F = 7.0\%$  larger than the bulk Si(004) atomic layer spacing. For such an ideal coherently strained film with  $N$  completely occupied Ge layers, the Fourier component introduced in Eq. (1) would be

$$\mathcal{F}_H = \frac{1}{N} \sum_{n=0}^{N-1} \exp[2\pi i(n\delta_H^F + \delta_H^I)]. \quad (3)$$

Where  $\delta_H^F$  is the fractional increase of the  $d$  spacing of the film ( $d_H^F$ ) relative to the  $d$  spacing of the substrate ( $d_H^S$ ).  $\delta_H^I$  is the offset for the first epitaxial layer. We will assume that  $\delta_H^I = \frac{1}{2}\delta_H^F$  based on the difference in length between Si-Ge bonds and Ge-Ge bonds. From this equation, one can solve for the elasticity theory predicted values for  $f_H$  and  $P_H$ ,

$$f_H = C |\mathcal{F}_H| D_H = C \frac{\sin(N\pi\delta_H^F)}{N \sin(\pi\delta_H^F)} D_H, \quad (4)$$

$$P_H = \frac{1}{2\pi} \arg(\mathcal{F}_H) = \frac{(N-1)}{2} \delta_H^F + \delta_H^I. \quad (5)$$

In Eq. (4),  $D_H$  is the Debye-Waller factor and  $C$  is the fraction of atoms in the predicted positions (ordered fraction). By combining fundamental and higher-order Bragg reflection XSW results it is possible to further decompose  $f_H$  into its constituent parts.<sup>26,27</sup> In our case, we are able to separate out the ordered fraction ( $C$ ) and the Debye-Waller factor ( $D_H$ ) in Eq. (4) by performing (004) and (008) XSW measurements on the same samples. The Debye-Waller factor can be described as follows:

$$D_H = \exp\left(-2\pi^2 \frac{\langle u_H^2 \rangle}{d_H^2}\right). \quad (6)$$

$\langle u_H^2 \rangle^{1/2}$  is the rms thermal vibrational amplitude of the Ge atoms. Figure 2 shows the (a) (004) and (b) (008) XSW measurement on the same sample (note that  $P_{008} \cong 2P_{004}$ , as expected). Using the XSW measured values for coherent fractions from the two measurements, we directly determine the Ge vibrational amplitude to be  $\langle u_{001}^2 \rangle^{1/2} = 0.08 \pm 0.02$  Å, which is close to values for bulk Ge (0.068) and bulk Si (0.063). The measured vibrational amplitude corresponds to a  $D_{004}$  of 0.94. Assuming an isotropic Ge vibrational amplitude, this measured value of  $\langle u_{hkl}^2 \rangle^{1/2} = 0.08$  Å is used to eliminate  $D_H$  as an unknown in Eq. (4).

Figure 3 shows the measured and predicted (004) and (022) coherent fraction and coherent position values for the samples grown with Te. [The predicted values come from Eqs. (4) and (5) with  $C=1$ ] Notice that for Ge thickness below 9 ML our XSW coherent fractions and coherent positions agree well with values predicted from elasticity theory for pseudomorphic growth. This is in contrast to samples grown without a surfactant (as will be discussed later with

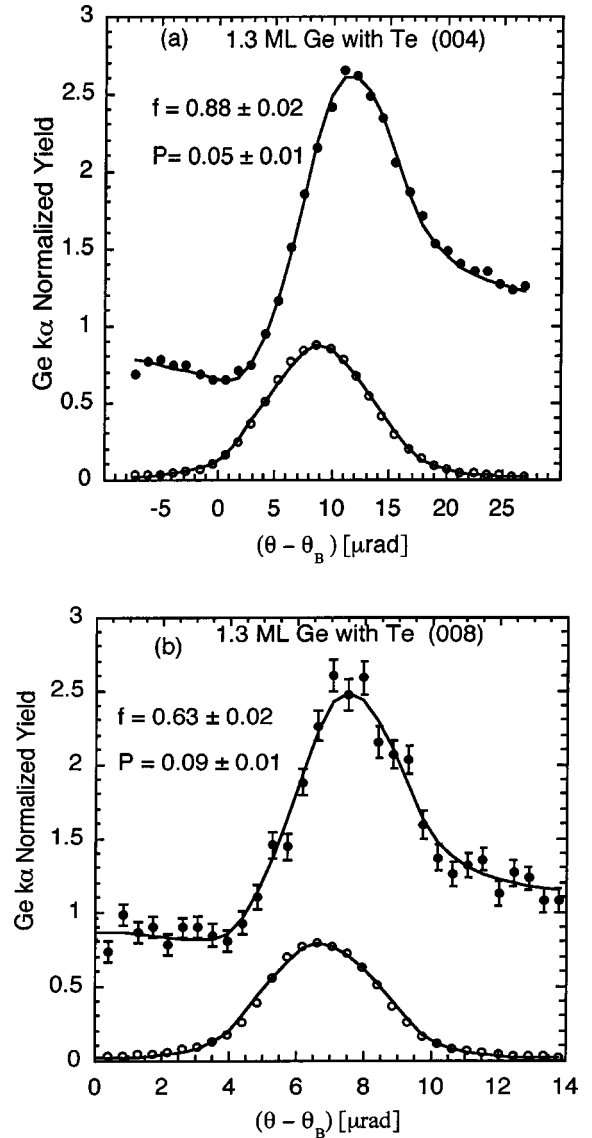


FIG. 2. The XSW experimental and theoretical angular dependency of the Ge  $K\alpha$  fluorescence yield and reflectivity collected while scanning in energy through (a) the Si(004) and (b) the Si (008) reflection. The Si/Ge/Si(001) sample has 1.3 ML Ge with Te as a surfactant.  $E_{\gamma}=12.2$  keV.

reference to Table I). To interpret the results in Fig. 3, we must consider the factor for ordered fraction ( $C$ ) in Eq. (4). For the case of  $C=1$ , all Ge atoms would be occupying “ideal” ordered tetragonal lattice positions. “Ideal” in this interpretation would pertain to positions predicted by the simple elasticity model. The tendency for Ge to segregate and form islands as a means of stress relaxation has the effect of creating a larger distribution of Ge positions and thus lowering the ordered fraction. In addition, the introduction of defects such as vacancies, dislocations, or twin boundaries would lower this value as well. For our 10.2 ML sample grown with Te, we believe that the introduction of such defects, as a means of strain relaxation, is responsible for the deviation of  $f_H$  and  $P_H$  values from elasticity theory predictions.

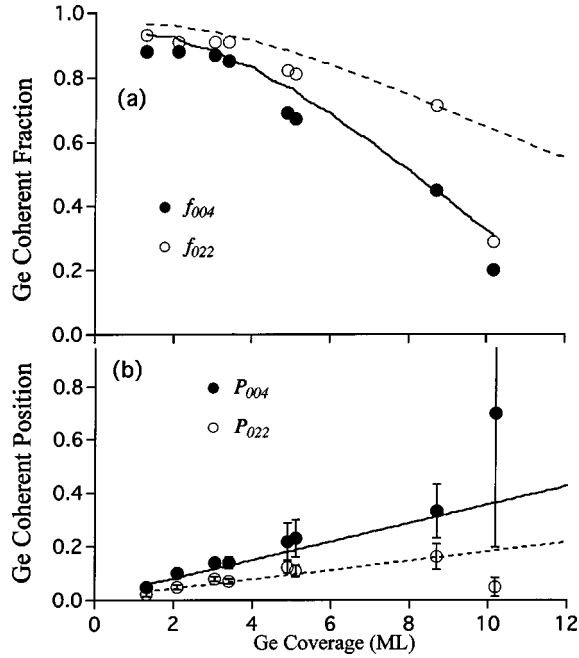


FIG. 3. The XSW measured (circles) and calculated (lines) values for (a) Ge coherent fraction and (b) Ge coherent position vs Ge coverage for Si/Ge/Si(001) samples with Te. The (004) solid line and (022) dashed line calculations are based on the elasticity theory. The calculated  $f_H$  values include a Debye-Waller correction and assume  $C = 1$ .

For comparison, we list calculated values for ordered fraction ( $C$ ), coherent position, and coherent fraction, for selected samples grown with and without surfactant in Table I. For the samples grown with surfactant,  $C$  is calculated from Eq. (5). In this equation,  $|\mathcal{F}_H|$  is calculated assuming that the epitaxial growth follows elasticity theory and  $D_H$  and  $f_H$  are measured quantities. For the samples grown without surfactant, the simple elasticity theory model is insufficient. For example, for the 3.7 ML sample grown without surfactant, it is expected that 3D growth will have initiated. This explains the relatively low coherent fraction for this sample ( $f_{004} = 0.37$ ). Notice that the two non-SME samples below the critical thickness have  $f_H$  and  $P_H$  values that are significantly less than those predicted by the ideal model in Fig. 3. This does not imply that the films are not pseudomorphic, but rather this indicates that there is significant diffusion of

TABLE I. (004) and (022) XSW measured coherent fraction and coherent position for samples without and with Te as a surfactant. The ordered fraction ( $C$ ) is included for samples with Te as determined from Eq. (4).

$\Theta$ (ML)	SME	$f_{004}$	$P_{004}$	$C$	$f_{002}$	$P_{022}$
1.3	√	0.88	0.05	0.95	0.93	0.02
3.4	√	0.87	0.14	1.00	0.91	0.07
5.1	√	0.67	0.23	0.90	0.84	0.11
1.1		0.72	0.03		0.74	0.01
2.7		0.67	0.05		0.65	0.04
3.7		0.37	0.23			0.12

Ge into the Si cap layer. This phenomenon has been well documented for Si/Ge (Refs. 28, 29) as it has been observed that a certain fraction of the Ge layer diffuses into, and even on top of the Si capping layer at growth temperatures between 350 and 650 °C. It has also been reported that Ge can diffuse into the subsurface Si layers.<sup>30</sup> It is therefore reasonable to assume that in addition to positions in the Ge epitaxial layer, Ge is occupying Si substitutional positions both below the Si surface and in the Si capping layer. The Ge that segregates to the surface of the Si cap will have oxidized into an amorphous phase. It is evident that there will be a wide range of Ge positions for the samples without Te, therefore, rendering the calculation of  $\mathcal{F}_H$  quite complicated. For these samples we could not include a value for  $C$  in Table I.

We have also recently performed a similar study, using Bi as the surfactant.<sup>31</sup> In this study, the Ge coherent positions for the samples grown with Bi agreed well with elasticity theory. However, the coherent fractions for samples above the critical thickness were markedly lower. This implies that like Te, Bi has the effect of preventing interdiffusion between Si and Ge layers during growth. However, Te is much more effective in preventing the formation of defects for Ge thin films above the critical thickness. Perhaps the most important prerequisite for a surfactant to be effective is to have a low activation energy for adatom site exchange. By providing a strong driving force for incorporation of Ge atoms into the epilayer, the surface diffusion of the Ge adatoms is reduced and defects such as vacancies or clusters are less likely to nucleate. This site-exchange property is strongly linked to the surface structure of the surfactant on both Si(001) and Ge(001). Group V elements dimerize on the surface of Si(001) and Ge(001) and passivate the surface, but leave behind a lone pair electron orbital. This is in contrast to group VI elements, such as Te, that bond at bridge sites on Si(001) and Ge(001) and render the outer electron shell of the surface atoms closed. While the group V  $2 \times 1$  terminated surface is more stable than is the clean Si or Ge surface, it most likely has a higher surface free energy than the Te-terminated  $1 \times 1$  surface. From this perspective, one can surmise how Te as a surfactant should be more effective than the group V surfactants and, therefore, it is evident why Ge layers grown with Te have a higher coherent fraction than those grown with Bi as a surfactant.

GIXD measurements at the APS DND-CAT were used to measure the in-plane lattice parameters for the specific purpose of distinguishing between the strained and relaxed Ge films. Figure 4 shows in-plane  $H$ - $K$  scans at  $L=0.03$  for three different samples. (Each had a 70 Å thick Si cap.) At this grazing incidence condition the scattering depth is  $\Lambda \approx 900$  Å and thus the in-plane scattering is sensitive to the structure of the Si cap, Ge buried layer, and Si substrate. For the 9 ML sample grown without a surfactant, a peak is present at  $H=K=1.93$ . This is close to the expected  $H=K=1.92$  position for a pure Ge bulk lattice constant, implying that there exists relaxed Ge in the heterostructure. The 9 ML sample that was grown with Te as a surfactant shows no feature at this  $H, K$  value. This indicates that the Ge epilayer is strained with an in-plane lattice constant constrained to that of Si(001). Figure 5 contains data for the same samples

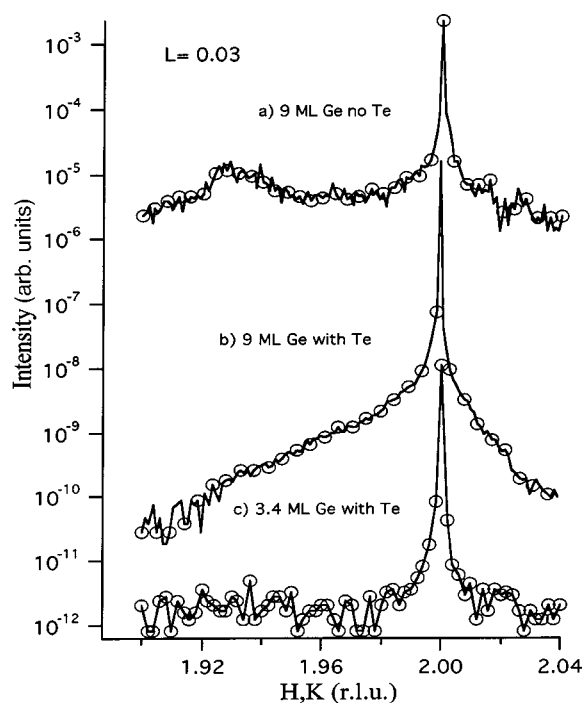


FIG. 4. GIXD  $H,K$  scans through the Si ( $2\ 2\ L=0.03$ ) peak for Si/Ge/Si(001) samples with (a) 9 ML Ge with Te as a surfactant, (b) 9 ML Ge with no Te, and (c) 3.4 ML Ge with Te as a surfactant.

in a more grazing incidence condition ( $L=0.01$ ). For this condition the calculated scattering depth is reduced to  $\Lambda \approx 20$  Å where only the Si capping layer should be probed. The peak at  $H=K=1.93$  for the 9 ML sample without surfactant is again apparent in this scan, implying a rough morphology with Ge islands in this sample. The calculation of  $\Lambda \approx 20$  Å assumes planar interfaces between Si and Ge. For the samples grown without Te, the Si cap/Ge interface is likely to be rough which will increase the scattering depth from our calculated value. In order to more thoroughly characterize the crystallinity of the epitaxial layers and to investigate the interface roughness, we have performed low-angle reflectivity and crystal truncation rod measurements on the samples in this study.<sup>32</sup> These results confirm that samples grown with Te have sharper interfaces with lower roughness (detailed analysis of these measurements will appear elsewhere).

### CONCLUSIONS

The efficacy of Te as a surfactant in growing Si/Ge/Si(001) heterostructures has been verified using XSW and GIXD. For samples that were grown with Te, the 2D growth

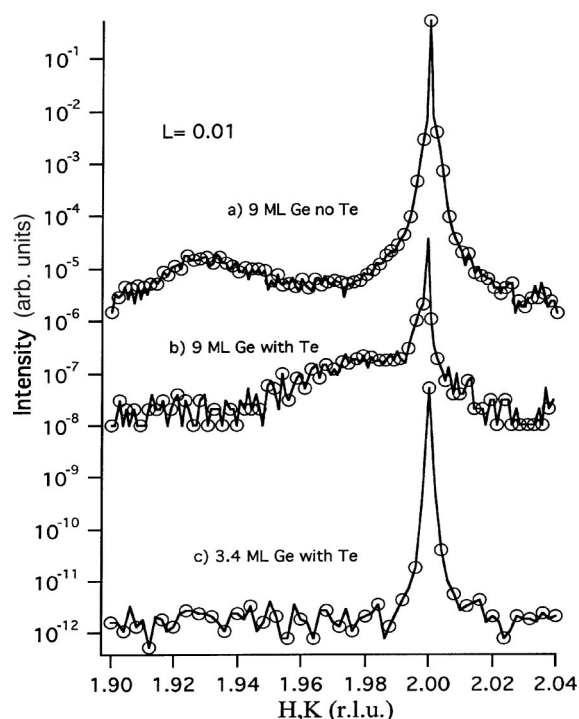


FIG. 5. GIXD  $H,K$  scans through the Si ( $2\ 2\ L=0.01$ ) peak for Si/Ge/Si(001) samples with (a) 9 ML Ge with Te as a surfactant, (b) 9 ML Ge with no Te, and (c) 3.4 ML Ge with Te as a surfactant.

regime was extended to at least 9 ML and the data agree with the continuum elasticity model for heteroepitaxy. Samples of similar thickness that were grown without a surfactant had a much lower coherent fraction than those grown with Te. This indicates intermixing and islanding for the Ge layers for these non-SME samples. The standing wave results were consistent with data obtained from grazing incidence diffraction where samples with Te proved to have a higher degree of crystallinity. The thermal vibration amplitude for Ge buried in Si were also determined, thus allowing us to more accurately calculate the Ge ordered fraction. Present investigations are underway to study thicker Ge films in order to determine the extent to which Te can be effective as a surfactant.

### ACKNOWLEDGMENTS

This work was supported by NSF-DMR under Contract Nos. DMR-9973436 and DMR-0076097 to NU and by DOE under Contract Nos. W-31-109-Eng-38 to ANL and DE-AC02-98CH10886 to BNL, and by the State of Illinois under Contract No. IBHE-HECA-NWU-96 to NU.

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