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Surfactant-mediated epitaxy of silicon germanium films on silicon (001) substrates

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ABSTRACT

We report on the surfactant-mediated epitaxy (SME) of $Si_{1-x}Ge_x$ films with $x = 0.23$–1 on Si(001) using antimony as surfactant. We observe a transition in strain relaxation at a critical composition $x_T = 0.58$-0.66. Above this value full relaxation is achieved by a network of full edge dislocation confined to the interface in analogy to SME of pure germanium on Si(001). 100 nm thick $Si_{1-x}Ge_x$ films with surface roughness values less than 1 nm and abrupt interfaces are obtained, as the surfactant reduces strain induced roughening and hinders interdiffusion.

1. Introduction

The growth mode of epitaxial $Si_{1-x}Ge_x$ films on Si is governed by lattice mismatch and thus film composition [1]. While layer-by-layer growth followed by strain relaxation via misfit dislocations is observed at small values of x, surface roughening and three-dimensional islanding of the elastically strained film prior to misfit dislocation nucleation are found at higher x. In the layer-by-layer regime, plastic relaxation relies on misfit dislocation nucleation at internal sources (e.g. defects) and dislocation multiplication; whereas misfit dislocation nucleation at the surface is the preferred strain relief mechanism for rough or islanded films (see e.g. [2] and references therein). The critical compositions x_{rough} and x_{3D} above which surface roughening or islanding occurs decrease with increasing growth temperature [1]. Surface roughening has been observed for compositions of $x_{rough} = 0.15$–0.3 at 833 K and $x_{rough} =$ 0.3–0.4 at 773 K, respectively [1]. A value of $x_{3D} = 0.5$–0.6 has been identified for growth on Si(001) at 873 K [3].

Different techniques have been developed to control the growth mode of $Si_{1-x}Ge_x$ films on Si. In addition to low temperature growth [4,5] or compositional grading [6], surfactants can be used to prevent islanding [7]. In surfactant-mediated epitaxy (SME) of pure Ge on Si at temperatures around 773 K surface roughening can be suppressed [8,9]. This has also been observed for relaxed $Si_{1-x}Ge_x$ films grown on compositionally graded or low temperature buffer layers [10,11]. In contrast, SME of Ge on Si with Sb as surfactant at temperatures above 873 K benefits from surfactant-controlled surface faceting yielding the abrupt formation of a regular dislocation network that compensates the lattice mismatch [12,13]. Thus, fully relaxed Ge films with low defect densities have been achieved on Si(001) [14] and Si(111) [15,16]. The impact of lattice mismatch on SME of $Si_{1-x}Ge_x$ on Si(111) at temperatures between 893 K and 973 K has been investigated by Kammler et al. [17]. In brief, the growth mode changes from layer-by-layer to surface faceting at $x = 0.6$. Only higher Ge contents ($0.6 \leq x \leq 1$) yield full strain compensation via a periodic misfit dislocation network.

In this paper, we investigate the growth of $Si_{1-x}Ge_x$ on Si(001) at 933 K using Sb as surfactant. We study the influence of lattice mismatch on the strain relaxation of $Si_{1-x}Ge_x$ films with $x = 0.23$–1 and thicknesses between 85 nm and 152 nm. Smooth relaxed 100 nm thick $Si_{1-x}Ge_x$ films with abrupt interfaces were obtained for $x > 0.66$ showing the potential of SME for the growth of thin relaxed buffer layers, which are indispensable for strain-engineered silicon–germanium multi-layer structures.

2. Experimental details

$Si_{1-x}Ge_x$ films with thicknesses between 85 nm and 152 nm and $x = 0.23$–1 were grown at 933 K on 100 mm diameter p-type (0.5–0.75 Ωcm) (001)-oriented Si wafers using molecular beam epitaxy (MBE). We applied an UV/ozone treatment followed by etching in 0.175% aqueous HF-solution until a hydrophobic surface was achieved prior to transfer into a VG 80 MBE-system with a base pressure below 2×10^{-8} Pa. After thermal treatment a clean 2 × 1 reconstructed surface was confirmed by reflection high-energy electron diffraction. Si and Ge were evaporated from electron beam sources at a total rate of 0.2 nm/s, while a constant surfactant (Sb) flux of 3×10^{13} cm^{-2} s^{-1} was supplied from an effusion cell. For comparison, nominally 100 nm thick films with $x = 0.31$–0.62 were also grown without a surfactant.

The sample morphology was inspected by scanning electron microscopy (SEM) using a ZEISS LEO DSM 982 Gemini operated at 5 kV. Layer thickness and surface roughness were determined by x-ray reflectometry

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Table 1
List of the $Si_{1-x}Ge_x$ samples grown in this study. Ge-contents x were obtained from HRXRD RSM measurements, layer thickness d was measured by XRR and surface roughness rms values r_{rms} by AFM.

Sample	x [%]	d [nm]	r_{rms} [nm]
90-01	23	96.2	0.2
69-08	35	84.9	0.3
69-14	44	85.0	0.4
69-09	52	89.9	0.3
70-25	58	96.9	0.3
69-13	66	94.4	0.4
70-04	75	97.3	0.6
69-02	77	95.4	0.7
70-05	100	152.0	0.4

(XRR) and atomic force microscopy (AFM), respectively. Film composition and degree of relaxation were obtained from high-resolution x-ray diffraction (HRXRD) reciprocal space maps (RSM) of asymmetrical reflections. X-ray analysis were carried out with a BRUKER D8 Discover diffractometer using Cu Kα_1 radiation, a Ge(220) Bartels-monochromator, and a channel cut analyzer, while AFM was performed on a PARK SCIENTIFIC M5 machine. Plan-view samples were prepared to investigate the microstructure of the $Si_{1-x}Ge_x$/Si interface by transmission electron microscopy (TEM) with a FEI Technai F20 microscope operated at 200 kV.

3. Results

All films have smooth surfaces and interfaces indicated by pronounced thickness oscillations in XRR measurements. AFM yields root-mean-square (rms) roughness values $r_{rms} < 1$ nm for all samples grown by SME (see Table 1). Samples grown without a surfactant show rms roughness values between 10 nm and 14 nm as illustrated in the scanning electron microscopy (SEM) images in Fig. 1.

Fig. 2 depicts the RSM of a sample with $x = 0.77$. In- and out-of-plane lattice constants close to the bulk value are found indicating almost full relaxation $R = 94\%$. The sample with $x = 0.23$ is coherently strained ($R = 0\%$) as the film thickness $d = 96$ nm is below the critical thickness for strain relaxation. For the other samples the degree of strain relaxation R extracted from RSM measurements changes with composition from $R = 52\%$ for $x = 0.35$ to $R \geq 94\%$ for $x \geq 0.66$ as shown in Fig. 3. The values depicted here are corrected for the tensile strain, which is induced in the cooling process after epitaxial growth due to the different thermal expansion coefficients of the $Si_{1-x}Ge_x$ film and the Si substrate [18]. The contribution of thermal strain to the measured in-plane strain increases linearly with germanium content. As R depends on the ratio of in-plane strain and misfit, the effect of thermal strain on R is given by the ratio of thermal strain and misfit. Applying Vegard's law to calculate the misfit between $Si_{1-x}Ge_x$ and Si, the ratio of thermal strain and misfit, and thus the correction to R, becomes independent of x. For the given experimental conditions a correction for R of −4.5% absolute is obtained.

The minute enhancement in the degree of strain relaxation observed in Fig. 3 for $x = 1$ can be explained considering the larger thickness of this film. The amount of residual elastic strain that can be stored in an epitaxial film is limited by the film thickness [19]. This maximum elastic strain is proportional to $1/d$ for residual strain values that are small compared to the lattice mismatch of the epitaxial system under consideration [19,20]. Thus, if the residual strain value or the degree of strain relaxation R_{d1}, respectively, is measured for a certain film thickness d_1, the residual strain value and hence, the degree of strain relaxation R_{d2} for a film thickness d_2 can be calculated

$$R_{d_2} = 1 - \frac{d_1}{d_2}\left(1 - R_{d_1}\right). \quad (1)$$

For a 95 nm thick Ge film a value of R = 97.6% is obtained from Eq. (1). This value has been included in Fig. 3 for comparison.

No broadening of the SiGe peak towards the substrate peak is observed in Fig. 2. This indicates an abrupt heterojunction between SiGe and Si. Rocking curve full width at half maximum (FWHM) values can be extracted from RSM measurements via line scans in azimuthal direction. The corresponding analysis yields a maximum value of 1700 arcsec for $x = 0.52$. The FWHM values decrease with increasing $x \geq 0.66$ down to 800 arcsec for pure Ge, while the degree of strain relaxation remains constant. This points to an improvement in structural perfection.

Fig. 4 shows plan-view TEM images of the film/substrate interface of samples with $x = 0.52$ (a) and $x = 0.77$ (b) taken under dark-field weak-beam conditions with $\vec{g}\,||(400)$. Two rectangular sets of misfit dislocations are visible as bright contrasts. For $x = 0.52$ a strong variation of misfit dislocation distance is found, whereas for $x = 0.77$ a periodic network of misfit dislocations is observed. In both cases the majority of misfit dislocations have been identified as edge type dislocations with Burger's vectors parallel to the interface applying the $\vec{g} \cdot \vec{b}$-extinction criterion under dark-field weak-beam imaging conditions with $\vec{g}\,||[220]$ and $\vec{g}\,||\left[2\bar{2}0\right]$. The misfit dislocation distance $D = 13$ nm obtained for the sample with $x = 0.77$ (Fig. 4(b)) is in perfect agreement with the expected value for $R = 94\%$ assuming relaxation via full edge dislocations. The overall picture revealed in TEM analysis is as follows: Regular arrays of misfit dislocations are exclusively found for Ge contents $x \geq 0.66$. The misfit dislocation distance corresponds to the measured degree of relaxation assuming relaxation via full edge dislocations and decreases with increasing x. $Si_{1-x}Ge_x$ layers containing less Ge exhibit unordered misfit dislocations. With decreasing Ge content an increased number of twins and stacking faults has been observed. The details of the defect structure and their role in the strain relaxation process are subject of ongoing investigations.

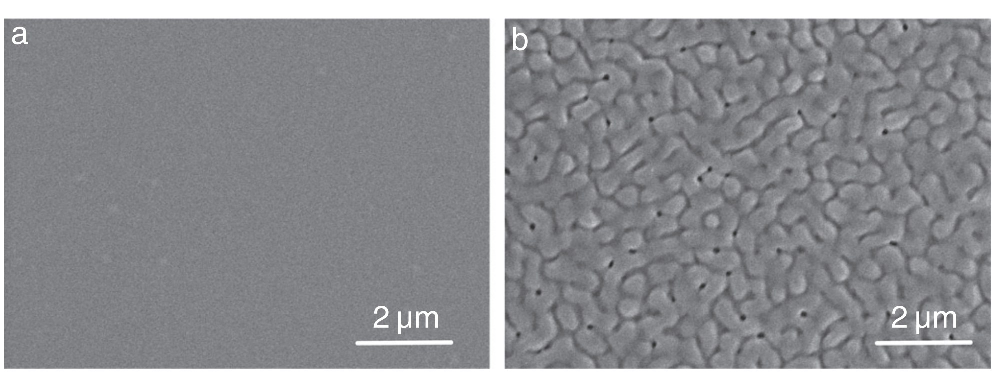

Fig. 1. SEM images of a 97 nm $Si_{0.23}Ge_{0.77}$ film grown with surfactant (a) shows a smooth surface ($r_{rms} = 0.4$ nm), while a 100 nm $Si_{0.34}Ge_{0.66}$ film grown without surfactant (b) has a pronounced surface roughness ($r_{rms} = 14$ nm).