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Effects of small specimen tilt and probe convergence angle on ADF-STEM image contrast of $\text{Si}_{0.8}\text{Ge}_{0.2}$ epitaxial strained layers on (100) Si

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ABSTRACT

The effects of specimen tilt and probe convergence angle on annular dark field (ADF) image contrast of $\text{Si}_{0.8}\text{Ge}_{0.2}$ heteroepitaxial strained layers on (100) Si were investigated in a 200 kV scanning transmission electron microscope (STEM) for a TEM specimen thickness of 195 nm. With 0.5 degrees of specimen tilt away from the exact $\langle 011 \rangle$ zone-axis orientation, the signal-to-noise level of atomic columns was significantly reduced for both $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si in high resolution ADF-STEM lattice images. When the specimen was tilted 0.5 degrees around the $\langle 011 \rangle$ axis, or the STEM probe convergence semiangle was reduced from 14.3 to 3.6 mrad, the ADF-STEM image intensity profiles across the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers changed significantly as compared to those obtained at the exact $\langle 011 \rangle$ zone axis orientation, and no longer reflected the composition changes occurring across the layer structure. Multislice image simulation results revealed that the misfit strain between the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers, and strain relaxation near the surface of the TEM specimen, were responsible for the observed changes in image intensity.

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1. Introduction

The introduction of strain into semiconductor epitaxial films can lead to enhanced performance in quantum well lasers, such as lower threshold currents and higher differential gain, due to strain induced changes in the electronic band structure [1]. However, the growth of perfect, planar, strained epitaxial films offers a number of interesting materials processing challenges in semiconductor device fabrication before the full benefits of strain may be realized. Conventional transmission electron microscopy (CTEM) has played an important role in characterizing strained epitaxial films. For example, CTEM studies of defects (dislocations, twins, stacking faults and cracks) and morphology helped to determine the strain relaxation mechanisms in SiGe/Si, InGaAsP/InP, GaAsN/GaAs films, which then facilitated optimization of growth methodology [2–5]. In recent years, annular dark field scanning transmission electron microscopy (ADF-STEM) has become a widely used and powerful technique for characterizing strained epitaxial layers due to the fact that ADF-STEM image contrast depends strongly on the atomic number Z of the scattering atoms in a simple Z^n form ($n=1.6$ – 1.9), which makes composition variation evident through a change in image intensity [6–10]. However, the existence of strain in the films and relaxation of strain

at the surfaces of the TEM specimen make interpretation of the measured intensity profile less straightforward. For example, atomic size mismatch strain induced reversed ADF-STEM image contrast between dilute semiconductor heteroepitaxial strained layers and substrates has been reported in GaNAs/GaAs [8,11,12] and SiC/Si systems [9]. In addition, a reduction in the ADF image intensity of InGaAs layers at both interfaces with adjacent GaAs layers due to strain relaxation has also been observed [10].

Channeling of the incident electrons along columns of atoms in crystalline specimens plays an important role in high resolution ADF-STEM imaging, where the axis of the incident electron probe was usually assumed to be exactly along the crystal zone-axis, despite the fact that a misalignment of the specimen of several milliradians from the targeted zone-axis orientation could be present and go unnoticed in microscope operation. Early reports in the literature claimed that ADF-STEM imaging was robust and insensitive to small changes in specimen tilt [13]. However, recent studies have revealed that ADF-STEM image contrast of crystals is sensitive to crystal orientation and a small tilt of the TEM specimen away from a low-index zone-axis orientation has been shown to reduce the visibility (signal-to-noise ratio) of atomic columns in high-resolution ADF-STEM images because electron channeling was affected [14–17]. The effects of specimen tilt on the ADF-STEM image contrast between epitaxial films and substrates have also been reported in InAsP/InP [7] and InGaAs/GaAs [10] heteroepitaxial systems. In order to better understand the effects of a 0.5 degree

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specimen tilt on ADF-STEM image contrast of epitaxial strained films on substrates, a study of the ADF-STEM image contrast dependence on specimen tilt in a $\text{Si}_{0.8}\text{Ge}_{0.2}/\text{Si}$ multiple quantum well structure has been performed. Furthermore, the effect of a change in the probe convergence angle on ADF-STEM image contrast was investigated.

2. Experimental procedure

The $\text{Si}_{0.8}\text{Ge}_{0.2}/\text{Si}$ sample consisted of four 40 nm thick $\text{Si}_{0.8}\text{Ge}_{0.2}$ strained layers separated by 40 nm thick Si spacer layers and was

Fig. 1. Illustration of the atomic arrangement of the Si and $\text{Si}_{0.8}\text{Ge}_{0.2}$ unit cell projected along the $\langle 011 \rangle$ zone-axis orientation.

grown by molecular beam epitaxy (MBE) on a Si (100) substrate. Details on the growth procedure have been previously reported [18]. High resolution X-ray diffraction was used to confirm that the quantum well structure was coherently strained as well as to determine the average Ge concentration in the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers.

Presented in Fig. 1 is an illustration of the atomic arrangement of Si and $\text{Si}_{0.8}\text{Ge}_{0.2}$ unit cells projected along the $\langle 011 \rangle$ zone-axis orientation. There was a misfit strain between the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers and the Si substrate due to the difference in lattice constants between the film and the substrate. The lattice constant of $\text{Si}_{0.8}\text{Ge}_{0.2}$ (0.5474 nm) is greater than that of Si (0.5431 nm), so the $\text{Si}_{0.8}\text{Ge}_{0.2}$ films were under biaxial compressive stress resulting in a tetragonal distortion of the $\text{Si}_{0.8}\text{Ge}_{0.2}$ lattice [9]. We defined a coordinate system in which the y-axis was parallel to the $\langle 100 \rangle$ growth direction and the plane containing the x- and z-axes was parallel to the film–substrate interface with the x- and z-axes along two orthogonal $\langle 011 \rangle$ directions (Fig. 1). After epitaxial growth, the lattice parameters of Si, using the unit cell presented in Fig. 1, remained at the bulk values of $b=0.5431$ nm and $a=c=d_{\langle 011 \rangle}=0.3841$ nm. For the strained $\text{Si}_{0.8}\text{Ge}_{0.2}$ film, the in-plane lattice parameters were constrained to match the Si substrates values, $a=c=0.3841$ nm. The out-of-plane lattice constant of the film, b , can be calculated from the linear elasticity theory and is given by $(1 - (2\nu f)/(1 - \nu))a$, which leads to the value $b=0.5511$ nm.

$\langle 011 \rangle$ cross-sectional TEM samples were prepared following standard dimpling and ion milling procedures. A 200 kV JEOL JEM-2100F TEM/STEM equipped with an ultra-high resolution pole piece ($C_s=0.5$ mm), a Fischione ADF detector (4–28 mm

Fig. 2. STEM probe size measurements: (a) image of probe formed using a 40 μm objective aperture (14.3 mrad convergence semiangle), (b) image of probe formed using a 10 μm objective aperture (3.6 mrad convergence semiangle) and (c) intensity line scans across the center of the two probes.

active diameter), a Gatan DigiScan and a Gatan Imaging Filter (GIF) Tridiem was used for this study. The theoretical optimum probe convergence semiangle and objective lens defocus were calculated to be 10.7 mrad and 26.6 nm, respectively, for this microscope [19]. Using the default set of the STEM objective apertures (corresponding to the conventional TEM condenser apertures) installed in the JEM-2100F, the two closest-to-optimum apertures had convergent semiangles of 14.3 mrad (40 μm aperture) and 3.6 mrad (10 μm aperture), and these two apertures were used in this study. The two STEM probes with 14.3 and 3.6 mrad convergence semiangles were reproduced in the TEM CBD (convergent beam diffraction) mode using the “Free Lens Control” function in the JEM-2100F and recorded using the GIF CCD camera as shown in Fig. 2(a) and (b). The line intensity profiles measured across the center of the probes are shown in Fig. 2(c). Both the 14.3 and 3.6 mrad probes displayed a Gaussian profile with full-widths at half-maxima (FWHM) of 0.26 nm and

0.36 nm, respectively. These probe sizes were greater than the minimum probe capabilities of STEM (~ 0.15 nm) since the beam current was increased to provide enough signal for ADF-STEM imaging at the highest ADF detector inner semiangle used (92 mrad). It is noted that the 14.3 mrad convergence semiangle forms the smaller of the two probes since it is closer to the optimum probe convergence semiangle of 10.7 mrad, and unless otherwise stated, all ADF-STEM images were acquired using this condition. The defocus value was chosen to achieve the highest contrast between the bright and dark spots in high magnification ADF-STEM lattice images. When recording the low magnification ADF-STEM images, the images were first focused at high magnification and then the magnification was lowered to the desired value.

Using the “Free Lens Control” function in the JEM-2100F, three ADF detector inner semiangles, 42 mrad, 67 mrad and 92 mrad, were obtained. The ADF detector inner semiangles were calibrated

Fig. 3. Definition of +x, -x, +y and -y tilts: (a) schematic diffraction pattern for Si at the $\langle 011 \rangle$ zone-axis orientation, and STEM diffraction patterns recorded using a GIF CCD camera of (b) $\langle 011 \rangle$ zone-axis, (c) after 0.5 degrees of +y tilt, (d) after 0.5 degrees of -y tilt, (e) after 0.5 degrees of +x tilt and (f) after 0.5 degrees of -x tilt.

by simultaneously recording $\langle 011 \rangle$ zone-axis Si diffraction patterns and the shadow of the inner edge of the ADF detector using the GIF CCD camera. The ADF detector outer collection semiangle, for all inner detector angles, was 175 mrad, the maximum angle accessible on the JEOL JEM-2100F with an ultra-high resolution pole piece [9]. Care was given to properly adjusting “brightness” and “contrast/gain” knobs of the ADF detector in order to obtain fairly accurate and meaningful image intensity measurements [20].

Since the $\text{Si}_{0.8}\text{Ge}_{0.2}$ film growth direction was along $\langle 100 \rangle$ and the interface between the strained $\text{Si}_{0.8}\text{Ge}_{0.2}$ film and Si substrate was parallel to the $\langle 011 \rangle$ direction (Fig. 1), the specimen was systematically tilted around the $\langle 100 \rangle$ (y-axis) and $\langle 011 \rangle$ (x-axis) directions to study the effect of the specimen tilt on ADF-STEM image contrast. When tilting the specimen ϕ degrees around $\langle 100 \rangle$ towards positive x or negative x , the tilts were defined as ϕ degrees of $+x$ or $-x$ tilt, respectively. Similarly, when the specimen was tilted ϕ degrees around $\langle 011 \rangle$ towards positive y or negative y , the tilts were defined as ϕ degrees of $+y$ or $-y$ tilt (Figs. 1 and 3a). The $\pm x$ and $\pm y$ tilts were achieved using the double-tilt holder in the orthogonally-driven goniometer while watching the diffraction pattern, and an example of how ϕ degrees of $+y$ tilt was obtained is described as follows. The specimen was first tilted to the $\langle 011 \rangle$ zone-axis orientation, and the diffraction pattern of the $\langle 011 \rangle$ zone-axis in the STEM mode was recorded using the GIF CCD camera (Fig. 3b). The angular readings of the goniometer at the $\langle 011 \rangle$ zone-axis were α_0 and β_0 for the X and Y axes, respectively. Using the live view of the $\langle 011 \rangle$ zone-axis pattern as a reference, the specimen was then tilted to the $+y$ tilt orientation by combining the X and Y tilts of the goniometer. The angular readings of the goniometer at the $+y$ tilt were α_1 and β_1 for the X and Y axes, and the total tilt ϕ

was given by the equation:

$$\cos(\phi) = \cos(\alpha_1 - \alpha_0)\cos(\beta_1 - \beta_0). \quad (1)$$

Figs. 3(c)–(f) are diffraction patterns after 0.5 degrees (8.7 mrad) of $+y$, $-y$, $+x$ and $-x$ tilts, respectively. In order to ease the specimen tilt for obtaining certain orientations and to minimize the error caused by operating the goniometer, the TEM specimen was loaded into the holder so that the interface (i.e., $\langle 011 \rangle$ direction) was nearly parallel to the X -axis of the goniometer and the superfine piezo stage was used. It is noted that the minimum tilt step reading of the goniometer was 0.1 degrees.

The thickness of the TEM sample was determined from the ratio of the plasmon to the zero-loss peaks in the electron energy loss spectrum. We choose a relatively thick sample area of ~ 195 nm for this study for two reasons. First, the sample was significantly bent in the thinnest regions, and it was impossible to uniformly orient the sample over a large area that included all four $\text{Si}_{0.8}\text{Ge}_{0.2}$ quantum well layers. Second, the HAADF image contrast between $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si is nearly independent of the sample thickness when the thickness is between 120 nm and 300 nm, as detailed in a previous study [9].

3. Observations

A high magnification ADF-STEM lattice image from one of the $\text{Si}_{0.8}\text{Ge}_{0.2}$ /Si interfaces recorded at the $\langle 011 \rangle$ zone-axis orientation is shown in Fig. 4(a). As expected, the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layer was much brighter than the Si layer, which is consistent with a Z -contrast interpretation since the average atomic number was greater for the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layer (17.6) than in the adjacent Si layer (14). Fig. 4(b)

Fig. 4. High magnification ADF-STEM images acquired using a 42 mrad ADF detector inner semiangle: (a) $\langle 011 \rangle$ zone-axis orientation, (b) after 0.5 degrees of $+y$ tilt and (c) after 0.5 degrees of $+x$ tilt.

and (c) are high magnification ADF-STEM lattice images acquired in the same region of the sample but with 0.5 degree $+y$ and $+x$ tilts, respectively. By tilting 0.5 degrees away from the zone-axis, the signal-to-noise level of the atomic columns was significantly reduced for both $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si, although the lattice resolution was preserved. This phenomenon, reported and discussed previously in other material systems, has been attributed to the effect of crystal tilt on electron channeling [14–17].

In order to study the effect of specimen tilt on the ADF-STEM image contrast between the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si quantum well layers and the Si substrate, low magnification images including all four $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers were recorded. An example of the intensity profiles across the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers obtained from an ADF-STEM image is shown in Fig. 5(a). The ADF-STEM image was obtained at the $\langle 011 \rangle$ zone-axis orientation with a 42 mrad detector inner semiangle. The central area of this image was the same region where the images in Fig. 4 were recorded. The intensity line profile was integrated over a width of 100 nm along the $\langle 100 \rangle$ growth direction and exhibited short range fluctuations in the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers, but was fairly uniform in the Si spacer layers. The former fluctuations point to the presence of variations in the Ge concentration in the growth direction and this phenomenon was discussed previously [9,18]. A comparison of the intensity profiles obtained at the $\langle 011 \rangle$ zone-axis orientation for three detector inner semiangles, 42 mrad, 67 mrad and 92 mrad, is presented in Fig. 5(b). The intensity profiles for all three detector angles are qualitatively very similar, with the absolute intensity decreasing with inner detector semiangle since fewer electrons were collected at higher scattering angles. In addition, there is an upward trend in the intensity towards the substrate, which is believed to be due to increasing specimen thickness in this direction.

The contrast (C) between the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers is defined as,

$$C = (I_{\text{SiGe}}/I_{\text{Si}}) - 1. \quad (2)$$

Fig. 5(c) is a plot of this average contrast as a function of the ADF-STEM detector semiangle and it is observed that the contrast increases with increasing inner detector angles, in agreement with an earlier study: in compositional contrast dominated SiGe/Si system, atomic scattering caused by elastic and strain contribute negatively to overall ADF-STEM image intensity [9].

Presented in Fig. 6 are the intensity profiles obtained from ADF-STEM images located at the same region as Fig. 5, but after the specimen has been tilted by 0.5 degrees in the $+y$ and $-y$ directions. With this level of specimen tilt, the intensity profiles across the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers changed appreciably. As compared with the image intensity recorded at the $\langle 011 \rangle$ zone-axis orientation (Fig. 5b) for a 0.5 degree $+y$ tilt, the ADF-STEM image intensity dropped at the start of each of four $\text{Si}_{0.8}\text{Ge}_{0.2}$ growth layers and then increased towards the end of the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layer growth along the film growth direction, while the intensity of Si spacer layers decreased along the film growth direction (Fig. 6b). The opposite behavior in the ADF-STEM image intensity was observed for the opposite tilt, a 0.5 degree $-y$ tilt (Fig. 6d). The intensity measurements obtained from inner detector semiangles of 42, 67 and 92 mrad all displayed similar trends in image contrast.

Due to the geometry of the quantum well structure, where the interfaces between layers are oriented horizontally in the images, $+y$ or $-y$ tilt of the specimen causes the interfacial plane to tilt in the same direction. This tilt breaks the symmetry of the projected quantum well structure in that the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers overlap with one another at the interfaces. However, in the case of specimen tilts in the $+x$ and $-x$ directions (see Figs. 1 and 3), the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers do not overlap at the interface and the projected quantum-well geometry in the direction of the electron beam is maintained. Therefore, it was

Fig. 5. ADF-STEM image taken at the $\langle 011 \rangle$ zone-axis orientation and associated intensity profiles: (a) ADF-STEM image acquired using a 42 mrad ADF detector inner semiangle with the intensity profile superimposed, (b) intensity profiles obtained from ADF-STEM images acquired at 42, 67 and 92 mrad detector inner semiangles, (c) image contrast plotted as a function of ADF detector semiangle.

expected, and confirmed by experiment, that $+x$ and $-x$ tilts of the specimen should lead to equivalent results and only the 0.5 degree $+x$ tilt results are presented in Fig. 7. Unlike the 0.5 degree $+y$ and $-y$ tilt results, the 0.5 degree $+x$ tilt (and $-x$ tilt by symmetry) line profile data resulted in no significant difference compared with the $\langle 011 \rangle$ zone-axis data (Fig. 5b) for all three ADF inner detector angles investigated.

Fig. 6. ADF-STEM images and intensity profiles after tilting the specimen in the $\pm y$ directions and arrows indicating intensity change trends along the growth direction: (a) ADF-STEM image recorded using a 42 mrad ADF detector inner semiangle after 0.5 degrees of $+y$ specimen tilt with intensity profile superimposed, (b) intensity profiles obtained from ADF-STEM images acquired using 42, 67 and 92 mrad detector inner semiangles after 0.5 degrees of $+y$ specimen tilt, (c) ADF-STEM image recorded using a 42 mrad ADF detector inner semiangle after 0.5 degrees of $-y$ specimen tilt with intensity profile superimposed, (d) intensity profiles obtained from ADF-STEM images acquired using 42, 67 and 92 mrad detector inner semiangles after 0.5 degrees of $-y$ specimen tilt.

Fig. 7. Intensity profiles obtained from an ADF-STEM image acquired after tilting the specimen 0.5 degrees in the $+x$ direction.

The effect of probe convergence angle on the intensity profiles across the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers was also studied. Presented in Fig. 8 are the results obtained from electron probes with 14.3 mrad

and 3.6 mrad convergence semiangles for an ADF detector inner semiangle of 42 mrad at the $\langle 011 \rangle$ zone-axis orientation. Fig. 8(a) is a plot based on the original image intensity, whereas the data in Fig. 8(b) has been scaled so that the maxima in the intensities of the two curves were aligned. The image recorded using the 14.3 mrad probe convergence semiangle had not only higher intensity, but also higher contrast between the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers (Eq. (2)). The intensity profiles obtained from $\langle 011 \rangle$ zone-axis oriented images using the 14.3 mrad convergence semiangle probe have proved to be consistent with the composition profiles across $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers determined by energy dispersive X-ray spectroscopy (EDX) and electron energy loss spectroscopy (EELS) [9]. However, when the 3.6 mrad probe convergence semiangle was used to form an image, the contrast between the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers was reduced, and more importantly, there were intensity minima at both interfaces between the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers resulting in an intensity profile that is not in agreement with the composition profile.

4. Simulations and discussion

The simulated image intensities were calculated using the multislice code of Kirkland [21], modified to run in parallel on a Beowulf computing cluster [22] using 22 processors. First, the elastic displacement fields arising from lattice misfit and surface-relaxation effects were modeled using the finite-element method

Fig. 8. Intensity profiles obtained from ADF-STEM images acquired at two different probe convergence semiangles, 14.3 mrad and 3.6 mrad: (a) original intensity profiles, and (b) intensities aligned with the maximum value.

and the software used was FlexPDE3 from PDE Solutions, Inc. [23]. Anisotropic elasticity theory was used and the elastic constants of Si [24] were assumed for all of the layers. The x and z dimensions were chosen to be 195 nm, the same thickness as the specimen used in the STEM measurements, and the y dimension was chosen to be 10,000 nm, a value much larger than the layer thickness so that all elastic strain effects occurred in the layers and not the substrate (thick substrate approximation). The finite-element grid of the quantum-well structure projected in the x – y plane along $z=97.5$ nm is presented in Fig. 9 where the elastic displacements have been increased by a factor of 50 to highlight the deformation at the surfaces of the structure. In this geometry, the electrons would be incident on the $x=0$ surface and exit through the $x=195$ nm surface. As expected, the larger lattice parameter of the $\text{Si}_{0.8}\text{Ge}_{0.2}$ (0.5474 nm) layers, as compared to the Si (0.5431 nm) layers and substrate, resulted in an outward displacement of the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers with respect to the Si layers. The largest displacements occurred near the surface of the sample where there were no physical constraints imposed. The x - and y -direction elastic-displacement fields are presented in Fig. 10. Although the displacements in the x -direction (in the direction of the electron beam) are about an order of magnitude larger than in the y -direction (perpendicular to electron beam), it is

Fig. 9. An x – z slice (through the center of the y dimension) of the grid used in the finite element simulation where the elastic displacements have been increased by a factor of 50. The yellow represents the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers and the blue represents the Si layers. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the y -direction displacements that have the dominant effect in STEM ADF images causing the electrons to de-channel out of the atomic columns [10].

The x , y , and z displacement data obtained from the finite-element simulations were used to adjust the positions of the atoms in a $195 \times 195 \times 500$ nm³ supercell containing the quantum-well layers and 180 nm of the Si substrate. This size of cell was prohibitively large for multislice calculations where a minimum scattering semiangle for the ADF detector of 175 mrad was required. Thus, the single, large supercell was separated into 500 smaller cells where the center of each was spaced 1 nm apart. For the simulations of the image intensities using the 14.3 mrad aperture, the dimensions of the smaller cells used in the multislice calculations were $x=3.46$ nm, $y=3.80$ nm and $z=195$ nm, which corresponded to $9 \times 7 \times 508$ of the unit cells presented in Fig. 1. Only the central cell, with x and y numberings of 5 and 4, respectively, was sampled to determine the intensity at a particular location. This size of unit cell was large enough so that edge effects resulting from the periodic boundary conditions of the FFTs used in the multislice calculations did not significantly impact the intensity at the center of the unit cell [25]. In addition, this choice of unit cell was small enough to permit a maximum scattering angle in the simulations of 225.2 mrad using a 1024×1024 pixel wave function, which exceeded the maximum angle used in the experimental images of 175 mrad [21]. The central cell was sampled in a 9×13 grid ensuring that the requirements of Shannon's sampling theorem were met as reported by Dywer [26]. The sampling parameters for the simulations of the image intensities involving the 3.6 mrad aperture were different from those used in the 14.3 mrad simulations in order to maintain a minimum of 10 pixels in reciprocal space that could be used to form the electron probe, as well as to maintain a maximum scattering angle of 225.2 mrad. Thus, the number of replicates of the unit cell in Fig. 1 was increased from 9×7 to 18×14 , the dimension of the wave function was increased from 1024×1024 pixels to 2048×2048 pixels, and since the electron probe is larger, the sampling within the central cell could be reduced from 9×13 points to 2×3 points while still satisfying the Shannon's sampling theorem. Note that the effects of the probe effective size were not included in current simulations. In order to simulate a line scan, the full multislice calculations were repeated 125 times at 4 nm intervals starting at the upper surface of the

Fig. 10. (a) x - and (b) z - elastic displacement fields simulated for the quantum well structure for a slice taken through the center of the y dimension. All values in nm.

sample and proceeding to the Si substrate. Furthermore, each location was repeated 8 times with slightly different atomic configurations and the intensities were averaged incoherently to account for thermal vibration effects of the atoms in the frozen phonon approximation [21,27,28]. A slice thickness of 0.384 nm and defocus of 26.6 nm were used in the simulations where the defocus value corresponded to the 'most compact probe' criteria given in [19]. In the $\text{Si}_{0.8}\text{Ge}_{0.2}$ film, 20% of the Si atoms were assumed to be randomly substituted by Ge atoms and the virtual alloy approximation was not used since it can lead to significant errors in the frozen-phonon approximation [29]. The remaining simulation parameters corresponded to experimental conditions: spherical aberration coefficient of 0.5 mm, electron accelerating potential of 200 kV and condenser apertures of 14.3 mrad and 3.6 mrad, as required.

Presented in Fig. 11 are simulated intensity profiles of ADF-STEM images when the sample was located at the $\langle 011 \rangle$ zone-axis orientation, tilted 0.5 degrees in the $+y$, $-y$ and $+x$ directions (a–d, respectively). These simulation results agreed very well with the corresponding experimental observations shown in Figs. 6 and 7. Not only does the intensity change at the interface and across the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers, but also the relative intensities obtained from the three different ADF detector

angles matched closely. Also included in Fig. 11 is a plot of contrast as a function of ADF detector semiangle for the simulation results obtained at the $\langle 011 \rangle$ zone axis orientation (Fig. 11e), which agreed well with the experimental results with the same trend of increasing image contrast with increasing ADF detector semiangle. The image contrast values determined from the simulation were within 5% of the contrast values obtained experimentally for all three ADF detector semiangles. This difference in image contrast between experiment and simulation may be attributed to factors not included in the model. For example, if the Ge atoms were randomly substituted within the Si lattice, it would be expected that multi-atom clusters of Ge would be present that would lead to potentially large localized strains resulting in increased non-Bragg scattering and thereby increasing the simulated contrast values [8]. In addition, the effects of any inhomogeneous response of the ADF detector were not included [30].

In order to further study the effect of misfit strain and strain relaxation in a TEM specimen on the intensity profiles of $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers on Si, a multislice simulation neglecting the effects of misfit strain and surface strain relaxation was performed for the 0.5 degree $+y$ tilt condition and the intensity line profiles are shown in Fig. 12. These intensity line profiles are similar to those obtained at the $\langle 011 \rangle$ zone-axis orientation with strain effects included and are significantly different from the y -tilt simulations that included strain. There were no obvious intensity drops at the $\text{Si}_{0.8}\text{Ge}_{0.2}$ /Si interfaces, or increase or decrease in intensity within the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers. Thus, the effects of misfit strain and surface strain relaxation have been confirmed as the sources of the observed intensity profiles across $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers after the specimen had undergone a small tilt. As discussed in Section 2, the misfit strain in $\text{Si}_{0.8}\text{Ge}_{0.2}$ films due to the difference in lattice parameter between $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si was accommodated mainly by a tetragonal distortion of the $\text{Si}_{0.8}\text{Ge}_{0.2}$ lattice, and this coherent tetragonal lattice distortion strain was relieved partially at the two TEM specimen free surfaces resulting in lattice plane bending in the first few nanometers of the specimen surfaces, as shown in Fig. 9. Grillo's study revealed that the bending of the lattice planes in the TEM specimen surface due to strain relaxation results in two ADF-STEM image intensity minima at the GaAs/InGaAs interfaces even when the specimen is at exact $\langle 011 \rangle$ zone axis, and these contrast depth at minima decreases with increase in the specimen thickness, and is negligible when the thickness is greater than 100 nm [10]. In agreement with Grillo's study, the intensity minimum was not observed in the current $\text{Si}_{0.8}\text{Ge}_{0.2}$ strained layers with a thickness of 195 nm when the specimen is at zone axis (Figs. 5b and 11a). While small levels of specimen tilt may locally offset the effects of plane bending at one of the surfaces, the overall effect of elastic strain relaxation at the surfaces of the specimen is adverse to electron channeling down the atomic columns. This results in the asymmetric intensity profile at the interface of $\text{Si}_{0.8}\text{Ge}_{0.2}$ /Si, in agreement with the experimental and simulation results. It should also be noted that opposite ADF-STEM image intensity changes are expected to occur within the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers after a tilt about the y axis since the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers have opposite signs of plane bending, that is, the atomic planes within $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers are bent outward with respect to the direction of the electron beam whereas the Si atomic planes bend inward (Fig. 10b).

The simulation results of the intensity profiles obtained at two different probe convergence semiangles of 14.3 mrad and 3.6 mrad at the $\langle 011 \rangle$ zone-axis orientation and 42 mrad ADF detector inner semiangle are displayed in Fig. 13. These simulations qualitatively reproduce two experimental observations shown in Fig. 8: (1) there is an intensity decrease at both edges of the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers for the image acquired at 3.6 mrad convergence semiangle, and (2) the contrast between $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si is higher for the image acquired at

Fig. 11. Simulated intensity profiles of ADF-STEM images acquired: (a) at $\langle 011 \rangle$ zone-axis orientation, (b) 0.5 degrees of +y tilt, (c) 0.5 degrees of -y tilt, (d) 0.5 degrees of +x tilt and (e) experimental and simulation image contrast plotted as a function of ADF detector semiangle for $\langle 011 \rangle$ zone axis results.

14.3 mrad convergence semiangle. Coupled with the strain relaxation at the interfaces, the much smaller probe convergent angle seems to cause a Bloch-wave redistribution wherein the effective probe intensity became localized over 1s states, and thus enhance the channeling at the both sides of the $\text{Si}_{0.8}\text{Ge}_{0.2}/\text{Si}$ interface, which could result in the intensity drop. Furthermore, it has been shown in a recent study that insufficient convergent semiangle of the incident electron beam can produce intensity that does not depend on the atomic number [31] and additional study is required in order to better understand the effect of probe convergence angle on the ADF-STEM image contrast between epitaxial strained layers and substrates.

5. Conclusions

In this study, we have shown that small specimen tilt and probe convergence angle changes can significantly alter the ADF-STEM image intensity profiles across strained $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers on (100) Si substrates. When the specimen was tilted 0.5 degrees around the

$\langle 011 \rangle$ axis towards the $\langle 100 \rangle$ film growth direction (+y tilt), the intensity dropped at the start of the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layer growth, and then increased towards the end of the $\text{Si}_{0.8}\text{Ge}_{0.2}$ layer growth while the intensity of Si spacer layers decreased along the film growth direction. The opposite ADF-STEM image intensity changes were observed for a 0.5 degree -y tilt. The changes remained qualitatively the same for ADF detector inner semiangles of 42, 67 and 92 mrad. The intensity profile across the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si interface was unchanged if the specimen was tilted by 0.5 degrees around the $\langle 100 \rangle$ axis. Multislice image simulations revealed that the misfit strain between the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si, and the relaxation of this strain at the TEM specimen surfaces were responsible for the observed intensity changes. When the probe convergence semiangle was changed from 14.3 to 3.6 mrad, a decrease in the intensity profiles at both interfaces between the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si in the ADF-STEM images was observed. We also observed that the signal-to-noise level of atomic columns was significantly reduced for both $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si in high resolution ADF-STEM lattice images with the specimen tilted by 0.5 degrees away from the exact $\langle 011 \rangle$ zone-axis orientation.

Fig. 12. Simulated intensity profiles of ADF-STEM images acquired after a 0.5 degree +y tilt of the specimen without accounting for the effects of misfit strain and surface strain relaxation.

Fig. 13. Simulated intensity profiles of ADF-STEM images acquired at the $\langle 011 \rangle$ zone-axis orientation with two different probe convergence semiangles.

At the TEM sample thickness of 195 nm, the intensity profiles of $\text{Si}_{0.8}\text{Ge}_{0.2}$ layers on (100) Si obtained from ADF-STEM images acquired at $\langle 011 \rangle$ zone-axis orientation with 14.3 mrad probe convergent semiangle agreed very well with composition profiles obtained by EDX and EELS [9]. The strain in the films and relaxation of strain at the surfaces of the TEM specimen did not appear to significantly change the atomic number sensitivity of the ADF-STEM image. However, a small TEM specimen tilt, as well as a change in the probe convergent angle, altered the intensity profiles at the $\text{Si}_{0.8}\text{Ge}_{0.2}$ /Si interfaces, as well as within the

$\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers, thus making the intensity profiles invalid for composition analysis across the $\text{Si}_{0.8}\text{Ge}_{0.2}$ and Si layers. In order for ADF-STEM to be a meaningful alternative method to quickly estimate concentration and composition fluctuation in strained films, it is vital to use a proper probe convergent angle and align the specimen to the exact zone-axis orientation.

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