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The structural deformations in the Si/SiGe system induced by thermal annealing

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Abstract The structural deformations in Si/SiGe system during thermal annealing were investigated by means of atomic force microscope (AFM) and high-resolution X-ray diffraction (HRXRD). The (004) rocking curve measurements showed that the obvious fringes of rocking curve obtained from pre-annealing sample were faded out gradually and disappeared completely with increasing the annealing temperature, which indicated that the abrupt Si/SiGe interface was destroyed gradually. The analyses of the peak broadening and relative position of the SiGe epilayer with respect to the Si substrate in high-resolution reciprocal space map (HRRSM) measurements described clearly the formation of mosaic structure in Si/SiGe system. The inner deformation induced the surface corrugate, known as crosshatch morphologies, which was analyzed by AFM measurements.

Introduction

Due to the importance of SiGe layers in electronic and optoelectronic applications, it is now broadly employed in CMOS, bipolar, optoelectronics, sensors, and a variety of other areas. To realize the utilization of SiGe, strain relaxed and smooth alloy layers are required. To date, several types of buffer layers have been used to grow relaxed SiGe layers having a low threading dislocation density, including compositionally graded $Si_{1-x}Ge_x(0<x<1)$ layers, $Si/Si_{1-x}Ge_x$ superlattice, graded short-period $(Si_mGe_n)_N$ superlattice and low-temperature grown Si(LT-Si) layers etc. [1–5]. But, during SiGe processing, the smooth surface and abrupt interface were often destroyed. The morphological stability of a thin epitaxial film is also a subject of considerable scientific and technological importance, and the intermixing in the Si/SiGe interface alters the interfacial properties and degrades the device performance. So in this report we pay our attentions to the study of structural deformations in Si/SiGe system during thermal annealing by means of atomic force microscope (AFM) and high-resolution X-ray diffraction (HRXRD). The structural deformations include mainly two categories: one concerns the dislocation-induced surface corrugation, known as cross-hatch morphologies, and the other one deals with the deformation inside the film, often referred to as mosaic structures. Chen et al. [6] studied the formation of cross-hatch during the growth of SiGe film. About the mosaicity, such as the defects inside the SiGe epitaxial layers, HRXRD has been proved to be a powerful technique, because any structural change will broaden the X-ray diffraction peaks of SiGe layer.

Experimental

All the samples were cut into pieces of approximately 25×25 mm^2 from a Si(001)/SiGe wafer and annealed in an ultra high vacuum chamber, the temperature range for annealing experiments is 600–900 °C. The thickness of SiGe layer, composition, and epitaxial growth quality of

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pre-annealing sample and their alterations after annealing can be obtained from rocking curve, symmetric (004) and asymmetric (113) reciprocal space maps performed on a Rigaku ATX-E high-resolution X-ray diffractometer (at a wavelength of 1.5406 Å), which is equipped with a four-crystal Ge(2 2 0) monochromator and a channel-cut Ge(2 2 0) analyzer. The independent variation of the two diffraction angles ω (between primary X-ray beam and the sample surface) and 2θ(between incident and diffracted X-ray beams) of the diffractometer provides the possibility of high resolution reciprocal space map(HRRSM). The surface morphologies were observed by atomic force microscope (AFM) (digital instruments, nanoscope-III) and the surface roughness was characterized by root-mean-squared (RMS) roughness of the AFM images. The structural deformation inside the film was determined by observing the peak width and the relative position of the SiGe epilayer with respect to the substrate using HRRSM.

Results and discussion

Pre-annealing sample

In order to obtain precise information on the structural deformations in Si/SiGe system, all the samples before annealing should be the same, including the composition, strain and size. The samples were cut from a Si(001)/SiGe wafer with diameter of 200 mm supplied by Hitachi Kokusai Electric Inc.. The Si(001)/SiGe wafer was deposited by CVD at 530 °C with the partial pressure of 5.2 Pa for SiH_4 and 0.26 Pa for GeH_4 in total pressure of 26 Pa. The rocking curve and asymmetric (113) reciprocal space map of the pre-annealing sample are shown in Fig. 1 (a, b), respectively. From (004) rocking curve, we find that the pre-annealing sample shows the well-defined thickness fringes suggesting the abrupt interface. The characteristic oscillation indicates the high epitaxial growth of SiGe on Si substrate, and makes it easy to determine the thickness of SiGe layer and materials composition by the comparison between measured and simulated X-ray rocking curve. The thickness and mole fraction of Ge are 283 nm and 0.192, respectively. The thickness and structure can also be verified by cross-sectional HRTEM, because we can measure the thickness of SiGe layer and observe the mismatch between SiGe layer and Si substrate from HRTEM images. We found that the thickness is also about 280 nm and there is no any extra half-plane of atoms at the interface and no misfit dislocation on SiGe top layer from the HRTEM image inset in Fig. 1(a). Moreover, the crystal and structural qualities of SiGe film on Si substrate can also be seen from an asymmetric (113) diffraction. A typical HRRSM around asymmetric (113) reflections from pre-annealing

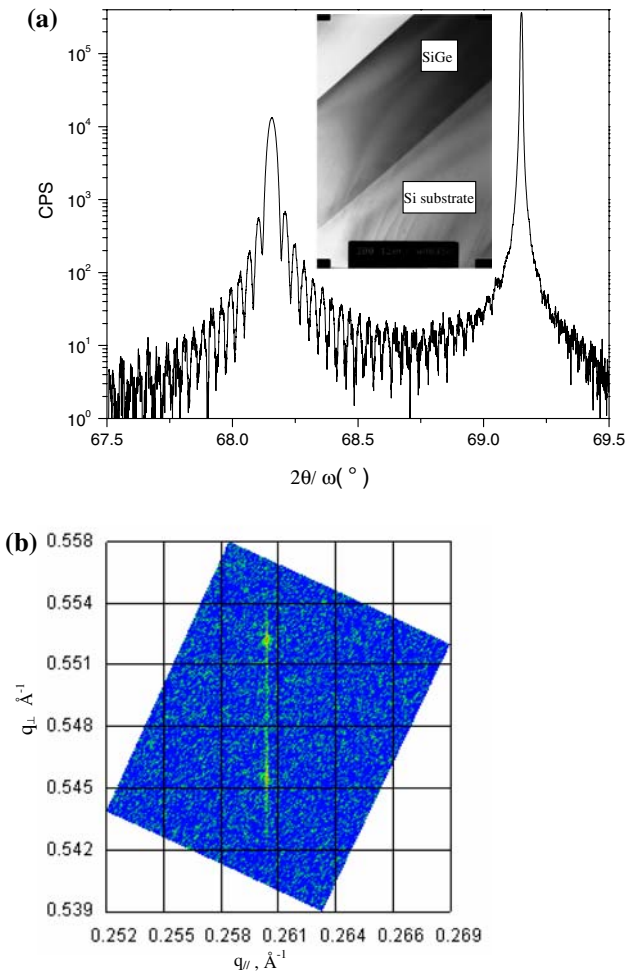

Fig. 1 (**a**) The (004) rocking curve of pre-annealing sample. Inset shows the cross-sectional HRTEM image of pre-annealing sample; (**b**) asymmetric (113) reciprocal space map of pre-annealing sample. The perfect raw materials are used for the experience

SiGe layer is shown in Fig. 1(b). The diffracted image is plotted as a function of the reciprocal lattice vector parallel $q_{//}$ and perpendicular $q_\perp$ to the surface. From the figure, we find that the scattering distributions of Si substrate and SiGe film in pre-annealing sample are very narrow in both ω and $2\theta/\omega$ scans, which shows that the substrate and layer scattering distributions are essentially perfectly aligned in the $q_{//}$ direction, that is, the alloy is commensurate with the substrate. The layer diffraction maps are also nearly symmetric, an indication of high crystalline quality and absence of mosaicity in SiGe layer.

Inner deformation

Figure 2 shows the typical (004) rocking curves of samples annealed for 30 min at different temperatures. There are some interesting features in these curves. For the samples annealed at low temperature, the oscillation of rocking

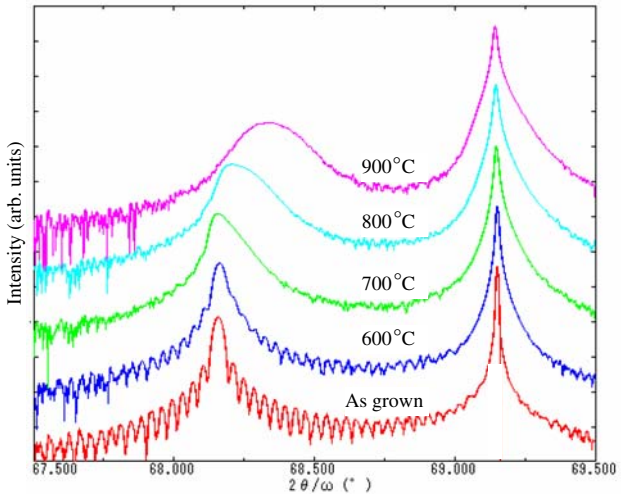

Fig. 2 The rocking curves of samples annealed 30 min at different temperatures. On increasing the annealing temperature, the oscillations are faded out gradually and disappeared completely at high temperature, and the center positions of SiGe(004) peaks gradually move toward that of Si(004)

curves is very clear. By contrast, there is no oscillation in the rocking curves of the samples annealed at high temperature, which means that the obvious fringes of rocking curve obtained from pre-annealing sample faded out gradually and disappeared completely with increasing the annealing temperature, indicating the interface was broadened gradually due to the deformation in the Si/SiGe system. The angular separation between the SiGe and Si peaks gradually decreased and the peak of SiGe moved toward that of Si substrate, suggesting a high temperature promoted the structural deformation at interface related to the strain relaxation. We also find that the Full-Width at Half-Maximum (FWHM) changed with different annealing temperatures and times, indicating the degradation of the interface was caused by the generation of dislocation. Moreover, the intensity of SiGe peak decays gradually with increasing the annealing temperature. The interdiffusivity can be calculated from the decay of the integrated intensity of the SiGe peak as a function of annealing time at different temperatures. Here, we will focus our attentions on analyzing the mosaic structures in the films. To show these features in the rocking curves more clearly, the two-dimensional distribution of the X-ray scattering intensity around the (004) points of Si and SiGe has been recorded by making a series of $2\theta/\omega$ scans while changing the ω angle gradually. Figure 3 shows the measured (004) reciprocal space maps of five representative samples, where (a) pre-annealing sample and annealed at (b) 600 °C; (c) 700 °C; (d) 800 °C; (e) 900 °C, respectively. The diffraction intensities are plotted with a logarithmic scale as functions of the reciprocal space vector components, $q_{//}$ and $q_{\perp}$. Since the SiGe alloy layer has larger lattice constant than Si, its corresponding peak appears at smaller $2\theta/\omega$ position (lower part of the map). With increasing the annealing temperature, the distribution of SiGe moves toward that of Si. The separation of the points of Si and SiGe is apparently smaller for the samples annealed at high temperature due to its higher degree of strain relaxation. Nonetheless, the diffraction points of the SiGe alloy layers are different in shape. For pre-annealing sample, the diffraction peaks of Si substrate and SiGe film are near point-like shape, namely there are no spread in both ω and $2\theta/\omega$ scans, indicating the uniformity of Ge composition and defects free in SiGe top layer. With increasing annealing temperature, the scattering distributions of Si substrate and SiGe layer spread gradually and the spread in $2\theta/\omega$ scan direction is wider than that in ω scan direction. The diffraction peaks were broadened by nonuniformity in the spacing of the lattice planes due to the inner deformation caused by the thermal annealing.

In order to analyze the inner deformation before and after annealing, we also studied HRRSMs around asymmetric (113) reflections. Figure 4 shows the (113) HRRSMs of four samples annealed at (a) 600 °C; (b) 700 °C; (c) 800 °C; (d) 900 °C, respectively. The diffraction intensities are also plotted with a logarithmic scale as functions of the reciprocal space vector components, $q_{//}$ and $q_{\perp}$. The inner fan-shaped quadrangle is the two axes scan region. The trendy of the distribution scattering of the peaks of Si substrate and SiGe layer in (113) diffraction is the same as that of (004) diffraction. But we find that after annealing, the position of the (113) SiGe layer peak is shifted toward the $2\theta/\omega$ axis from the center positions of SiGe and Si reciprocal lattice points of pre-annealing sample almost perfectly aligned in the $q_{\perp}$ direction, indicating the increase of strain relaxation in [110] direction. The center position of SiGe layer changed as a function of annealing temperature, which indicate the change of the out-of-plane lattice constant and in-plane lattice constant, corresponding to the strain relaxation in [001] and [110] directions. The calculation methods about the lattice constant and strain relaxation were described in detail in reference [7]. Apart from the calculations of lattice constant and strain relaxation, many papers discuss the reciprocal space map in qualitatively but quantitatively. Godwod et al. [8] introduced the mathematical analysis of the high-resolution reciprocal space map. The structural deformation inside the SiGe film can be determined quantitatively by measuring the peak width and the degree of the tilt of the epilayer with respect to the substrate using HRRSM. In the case of high dislocation density systems such as GaAs/Si, ZnSe/GaAs, InSb/GaAs etc., and the dislocation density can be evaluated according to the broadening in $2\theta/\omega$ and ω directions, however the evaluation for Si/SiGe system is under the consideration.

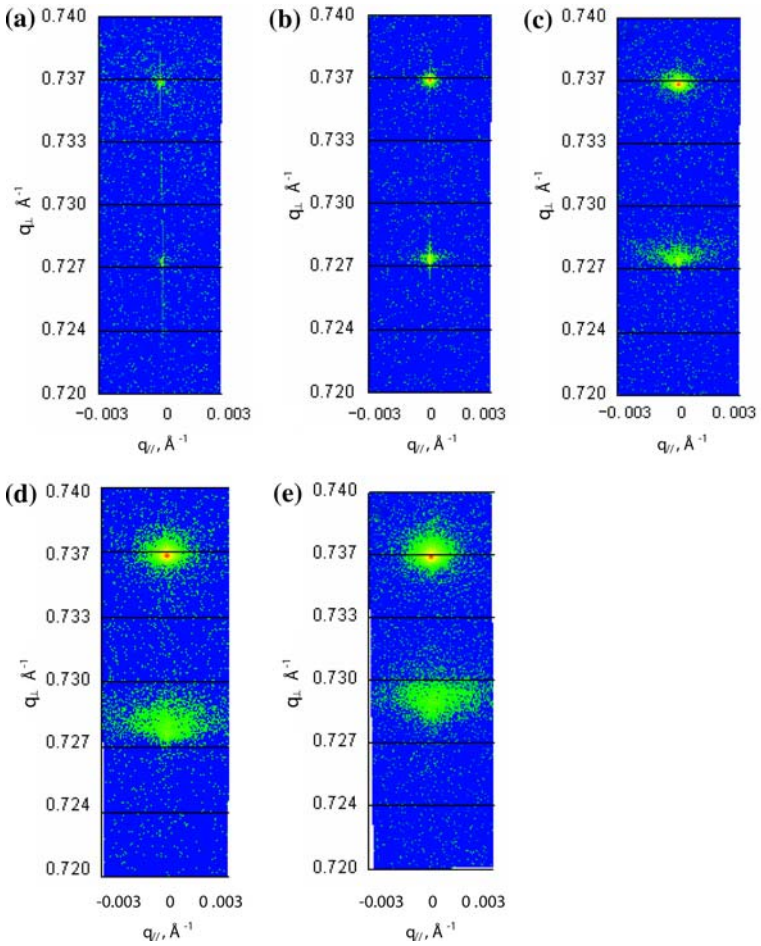

Fig. 3 (004) HRRSMs of samples annealed 30 min at different temperatures (**a**) pre-annealing sample; (**b**) 600 °C; (**b**) 700 °C; (**c**) 800 °C; (**d**) 900 °C

Surface deformation

The typical surface morphologies of the samples before and after annealing at different temperatures for 30 min measured by ex situ AFM are shown in Fig. 5. The scanning area is 20×20 μm^2 and the sides of these figures are parallel to [100] and [010] directions, respectively. The RMS roughness and the strain relaxation of the SiGe top layer are shown in Fig. 6. The surface of the pre-annealing sample grown on Si(001) showed no cross-hatch pattern on the surface with RMS <0.3 nm. The film remains still highly strained with relaxation(R) varying only from 0% to 2% as temperature increased to 600 °C from the growth temperature of pre-annealing sample of 530 °C. In this regime, the surface roughness is dominated by the formation of misfit-dislocation-induced surface ridges with the average feature of ridge-ridge separations d decrease with an increase in film relaxation. Pre-annealing sample, corresponding to a layer with R = 0%, the AFM image exhibits very smooth without any ridge; at 600 °C, corresponding to a layer with R = 2%, the AFM image exhibits a long, straight surface ridge which is aligned along one <110> direction. The average ridge-ridge separation is more than 15μm, indicating a few of misfit dislocations were caused at the Si/SiGe interface, which can also be confirmed by the narrower scattering distribution of SiGe peak in HRRSM. At 700 °C, corresponding to a layer with R = 5%, surface ridge increase in the observed area, and the crosshatch began to be formed which exhibit long, straight surface ridges are aligned along orthogonal <110> directions. The degree of film relaxation increases rapidly with annealing temperature T > 800 °C and the surface crosshatch morphology continues to develop as shown in AFM images consisting of both dislocation-induced steps and periodic arrays of ridges along 90°-rotated <110> directions. For the AFM image of a 800 °C annealing layer with R = 12%, the ridges are approximately symmetric in cross section and the average ridge-to-ridge space is 9 μm.

The relaxation of the 900 °C annealing sample is 25%, which increase rapidly from 12% of 800 °C annealing one. Both the average height and width of the ridges in this layer are larger than those of the 800 °C annealing film. From the AFM image, we obtain d = 2.8 μm, which is decreased obviously from 9 μm. And it indicates that coalescence is occurring in these highly relaxed layers. This is clearly observed in Fig. 5(e).

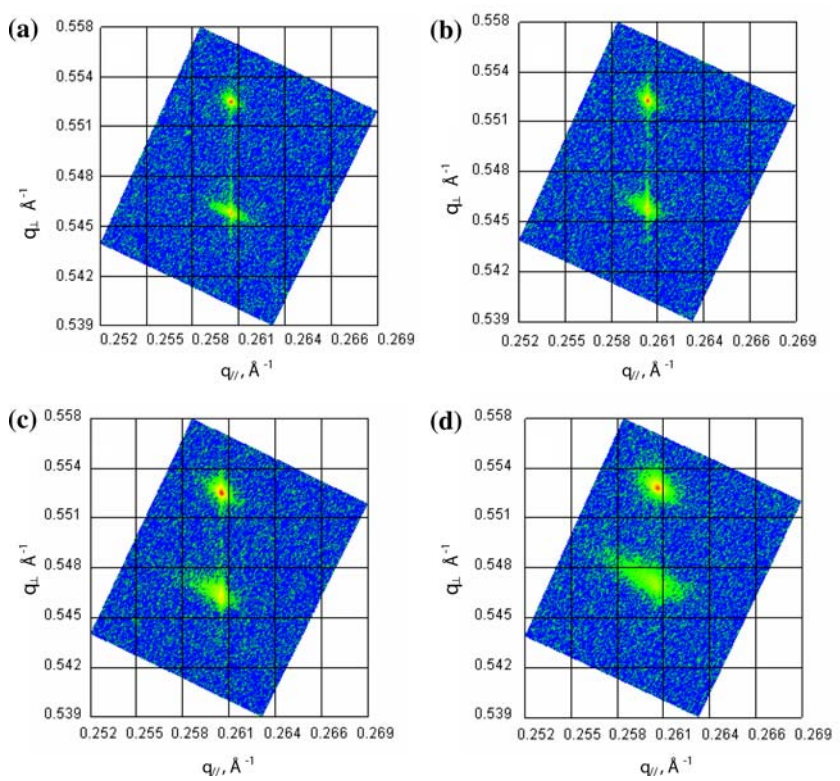

Fig. 4 (113) HRRSMs of samples annealed 30 min at different temperatures (**a**) 600 °C; (**b**)700 °C; (**c**) 800 °C; (**d**) 900 °C

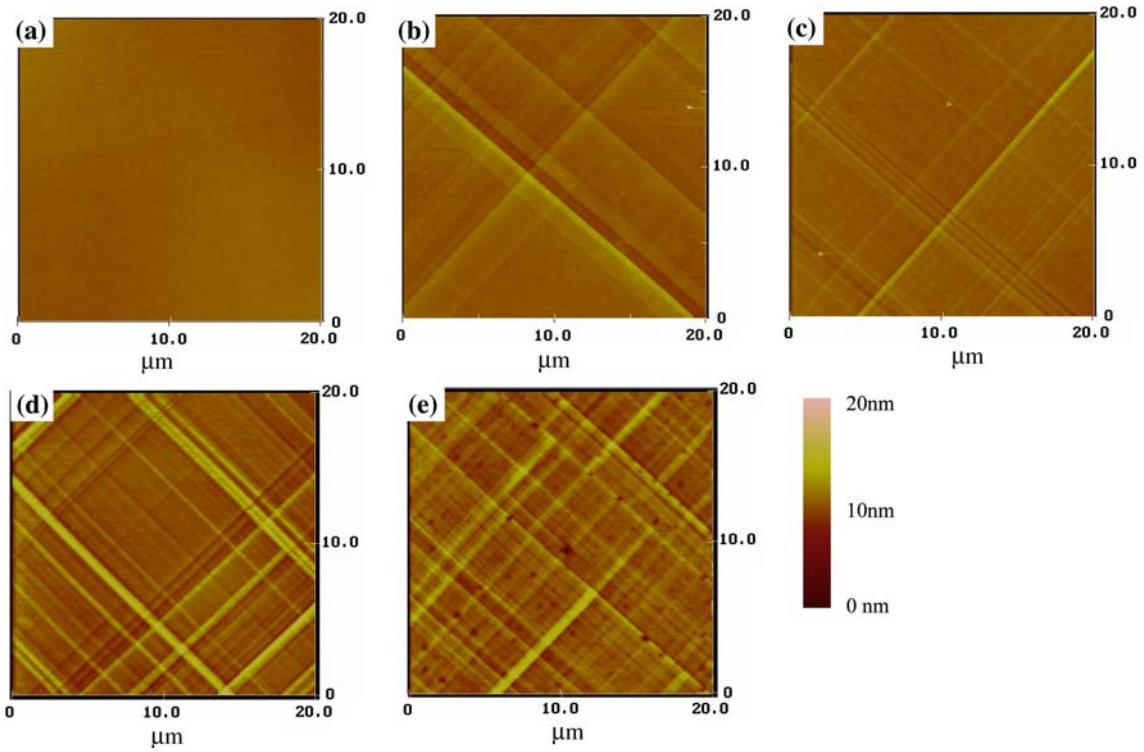

Fig. 5 AFM images of surface morphologies of samples annealed at different temperatures (**a**) pre-annealing sample; (**b**) 600 °C; (**c**) 700 °C; (**d**) 800 °C; (**e**) 900 °C

The deformations at the Si/SiGe interface induced by the thermal annealing provoke the structural change on the surface of SiGe top layer. Because of the lattice mismatch between SiGe layer and Si substrate, dislocations will occur in the interface during thermal annealing and propagate into the epilayer with increasing annealing temperature and

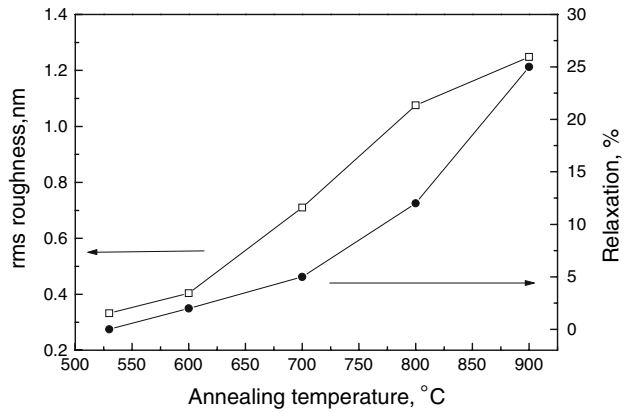

Fig. 6 The relaxation rate and the RMS roughness of the SiGe alloy layers as a function of the annealing temperature

prolonging annealing time, and epilayer/substrate interface where misfit segments are formed whose edge component relieves the strain accumulated in the layer. While gliding, the misfit segments interact with each other to create low energy sources for the introduction of new dislocations. During this movement, there is bunching of misfit segments that have the same Burgers vector. It is generally agreed that this bunching is the origin of the crosshatch. Most of the dislocations are mixed ones, where the crosshatch was caused by the misfit dislocation, at the same time a lot of threading dislocations will also appear on the surface, this is the reason for the pits on the surface (Fig. 5e).

Conclusion

Two kinds of structural deformations in thermally treated Si/SiGe films were systemically investigated by AFM and HRXRD. HRRSM measurements showed that the distribution scattering of Si and SiGe peaks broadened gradually due to the formation of mosaic structure in Si/SiGe systems with increasing the annealing temperature. The obvious fringes of rocking curve obtained from pre-annealing sample faded out gradually and disappeared completely with increasing the annealing temperature, indicating the interface was broadened gradually due to the deformations in the Si/SiGe system. The misfit dislocation caused by the inner deformation leaded to the formation of crosshatch pattern on the surface.

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