Surface and structural characterization of $Si_{1-x}Ge_x$/Si alloys and multi-quantum wells grown by gas source molecular beam epitaxy

L.-F. Zou,[1,*] S.E. Acosta-Ortíz,[1] L.E. Regalado,[2] LuXin Zou,[3] J. Sarabia-Torres,[4] and G.A. Pérez-Herrera[1]
[1]*Centro de Investigaciones en Óptica, A.C., Unidad Aguascalientes*
Juan de Montoro No. 207, Zona Centro, 20000 Aguascalientes, Ags., Mexico
[2]*Centro de Investigaciones en Óptica, A.C.*
Loma del Bosque No. 115, Loma del Campestre, 37000 Leon, Gto., Mexico
[3]*Computer Science Department, Zhongnan University for Nationalities*
Wuhan, Hubei 430074, China
[4]*Departamento de Eléctrica-Electrónica, Instituto Tecnológico de Aguascalientes*
Av. López Mateos y Av. Tecnológico, 20256 Aguascalientes, Ags., Mexico

Recibido el 20 de agosto de 1999; aceptado el 27 de junio de 2000

Gas source molecular beam epitaxy was employed to grow $Si_{1-x}Ge_x$ pseudomorphic layers at various germanium fractions, x, in the alloys with Si_2H_6 and GeH_4 as sources on (100) Si substrates. The structural characterization of $Si_{1-x}Ge_x$ pseudomorphic layers was investigated by double-crystal X-ray diffractometry, and surface characterization was studied using a scatterometer with angle resolved scattering for the first time. The signals with a scattering angle of $\pm 2^\circ$ around the specular direction for details of the polarization of light scattering were measured. Experimental results show that strained and partially relaxed $Si_{1-x}Ge_x$ alloys and $Si_{1-x}Ge_x$/Si multi-quantum wells (MQWs) present different scattering profiles, even if all of them display mirror-like surface morphology, which indicates that the surfaces of heterostructures are microrough to different degree with the samples grown under different growth conditions. We stress the importance of our results for the application of optical scattering technique as an optical probe for the study of surface microroughness in semiconductor heterostructures grown on lattice-mismatched surface.

Keywords: Microroughness; scattering; polarization; scatterometer; structural characterization

Se utilizó la técnica de epitaxia por haces moleculares con fuente de gas para crecer capas pseudomórficas de $Si_{1-x}Ge_x$ con diferentes fracciones x de germanio en las aleaciones, con Si_2H_6 y GeH_4 como fuentes, sobre substratos de Si (100). La caracterización estructural de las capas pseudomórficas de $Si_{1-x}Ge_x$ fue investigada por difractometría de rayos X de doble cristal y la caracterización de superficie se estudió utilizando por primera ocasión un esparcímetro con resolución angular. Se midieron las seales con un ángulo de esparcimiento de $\pm 2^\circ$ alrededor de la dirección especular para obtener detalles de la polarización de la luz esparcida. Los resultados experimentales demuestran que las aleaciones de $Si_{1-x}Ge_x$ parcialmente relajadas y las tensas, así como los pozos cuánticos múltiples de $Si_{1-x}Ge_x$/Si (MQWs), presentan diferentes perfiles de esparcimiento, aun cuando todas ellas muestren una morfología superficial tipo espejo, lo cual indica que las superficies de heteroestructuras tienen diferentes grados de micro-rugosidad para muestras crecidas bajo diferentes condiciones de crecimiento. Hacemos énfasis en la importancia de nuestros resultados para la aplicación de la técnica de esparcimiento como una prueba óptica para el estudio de micro-rugosidad superficial en heteroestructuras semiconductoras crecidas sobre superficies.

Descriptores: Micro-rugosidad; esparcida; polarización; esparcímetro; caracterización estructural

PACS: 68.55.Bd; 68.65.+g; 68.35.Bs

1. Introduction

It is well known that silicon and germanium are two of the most useful semiconductor materials. According to the Si-Ge phase diagram, the solid solution can be formed at any desired composition with an adjusted band gap. Recent advance in Si epitaxial technology has made it possible to grow pseudomorphic $Si_{1-x}Ge_x$ alloys with good crystallinity and electronic properties. The addition of Ge extends the wavelength range of Si, which allows an additional degree of freedom in band-gap engineering [1, 2]. High-quality surface of $Si_{1-x}Ge_x$ epilayers and high-quality $Si_{1-x}Ge_x$ alloys are essential for the realization of device structures for planar fabrication techniques in the semiconductor industry.

Because of the strain in $Si_{1-x}Ge_x$ epilayers, surface quality and structural characterization of epilayers have a closed connection with the growth parameters and growth modes in the heteroepitaxy. Double-crystal X-ray diffractometry (DCXRD) provides a rapid and nondestructive technique for evaluating the crystal perfection and strain in epitaxial films [3]. Light scattering has been shown to be a powerful diagnostic technique for characterizing surface quality [4, 5]. For optical surfaces even a small degree of roughness produces measurable light scattering. It was found that light scattered by microroughness has a high degree of polarization in all scattering directions [6]. But in contrast to the optical industry, light scattering has only recently been seriously considered as a source of microroughness characterization in the

semiconductor industry [4]. In order to characterize microroughness of semiconductor surface and optimize the design of electronic and optoelectronic devices, an accurate knowledge of the relation between the surface microroughness and the strain distribution in these heterostructures is essential.

In this paper we study the structural characterization of $Si_{1-x}Ge_x$ pseudomorphic layers by double-crystal X-ray diffractometry, and investigate surface characterization using a scatterometer with angle resolved scattering for the first time. The results show that surfaces have a closed connection with the strain distributions. Strained and partially relaxed $Si_{1-x}Ge_x$ alloys and $Si_{1-x}Ge_x$/Si multi-quantum wells (MQWs) present different scattering profiles, even if all of them display mirror-like surface morphology, which indicates that the surfaces of heterostructures are microrough to different degree for samples grown under different growth conditions. Our results are important for the application of optical scattering measurement to the study of surface microroughness in semiconductor heterostructures grown on lattice-mismatched surface.

2. Experiments

2.1. Gas source molecular beam epitaxy

Growth of $Si_{1-x}Ge_x$/Si alloys and multi-quantum wells was done in a modified MBE system by combining a gas-handling system, which has a vacuum load-lock entry and turbomolecular pump to produce pressure of about 2×10^{-4} Torr during growth [7, 8]. The growth chamber was provided with an ion pump which maintains a background vacuum 4×10^{-10} Torr. Pure Si_2H_6 and GeH_4 were selected as the source material. Each gas flow rate was controlled by precision mass flow controllers. The substrates used for the epitaxial depositions were (001)-oriented Si wafers, which were chemically cleaned using modified Shiraki' procedure [9]. Immediately prior to the loading, the substrate was dipped into the HF (2.5%) solution to remove the native oxide on the substrate surface. Then, the substrate was loaded into the growth chamber through the load-lock system. Prior to the growth, the substrate temperature was raised up to 900°C to desorb the remaining native oxide for 30 min, and the substrate temperature was brought down to the growth temperature. *In situ* reflection high-energy electron diffraction (RHEED) was used to monitor the surface crystallinity and smoothness. After all the growth, the surface of samples are mirror-smooth.

2.2. Double-crystal X-ray diffractometry

X-ray diffraction measurements were performed on Rigaku SLX-1AL double-crystal diffractometer with a Ge (400) monochromator. The X-ray source is $CuK\alpha_1$ (λ = 0.154 nm). In the present work the Bragg angles were measured in relation to the Si substrate (400) reflection at 34.56°. In this setup the X-ray beam spot size at the sample crystal was estimated to be 0.1 mm^2. Rocking curves are obtained by keeping the detector fixed at an angle 2θ with respect to the incoming X-ray beam and rocking the sample to vary θ by a small amount.

2.3. Scatterometer with angle-resolved scattering

Microroughness characterization of surface was investigated by a scatterometer with angle resolved scattering. The entire scatterometer assemble is constructed on a standard vibration isolated optical table. Stable He-Ne laser source with a polarized output 3 mW and a wavelength $\lambda = 632.8$ nm was used. A series of mirrors sent the polarized laser beam over a detector and through a half-wave plate. The beam was then reflected from a final mirror, passed through a polarizer, and was incident upon the surface of $Si_{1-x}Ge_x$/Si heterostructures. The half-wave plate enabled the incident polarization to be oriented as desired; the polarizer that followed removed any residual ellipticity of the polarization state. A slightly converging linearly polarized beam was sent toward the sample. The detection system, which consists of photomultiplier and polarization optical elements, was mounted on a 70 cm long arm and pointed toward the sample. The arm was rotated under computer control, and the sample mount was attached to a computer-controlled rotation stage fixed to the optical table so that the angle of incidence could be easily determined and modified by rotating the sample. The laser beam was modulated by a stabilized mechanical chopper, and detector signal was processed by a lock-in amplifier (Stanford Research System, SR530). The incident wave with an incident angle of 10° is linearly polarized in a direction orthogonal or parallel to the plane of incidence (s or p polarization, respectively) respect to the plane of incidence. The signals for the two polarization combinations, $s \rightarrow s$ and $s \rightarrow p$, with a scattering angle of $\pm 2°$ around the specular direction were measured because all samples show mirror-like surface morphology. Prior to the scattering measurements, the samples were cleaned chemically with a standard process to minimize the effect of contaminants. The incident beam was arbitrarily attenuated to a constant value for all the measurements. The intensity data were taken as a function of the scattering angle.

3. Results and discussion

Figure 1 shows the typical DCXRD rocking curves of $Si_{1-x}Ge_x$ alloys [Figs. 1a and 1b, FA312 and FA329, respectively] and $Si_{1-x}Ge_x$/Si MQWs [Fig. 1c, FE503]. For sample FA312, Fig. 1a, the peak amplitude, position and full width at half-maximum (FWHM) compare well with theoretical calculations as expected [10] based on elastic theory and dynamical theory [11, 12]. The Ge content of $Si_{1-x}Ge_x$ alloy is 0.11, corresponding thickness of $Si_{1-x}Ge_x$ epilayer is 170 nm, and the FWHM of SiGe peak is 180 arcsec, which indicates a high crystal quality of the $Si_{1-x}Ge_x$ alloy. For another $Si_{1-x}Ge_x$ alloy (Fig. 1b, FA329), the Ge content of $Si_{1-x}Ge_x$ alloy is also 0.11 that was obtained from the Ge

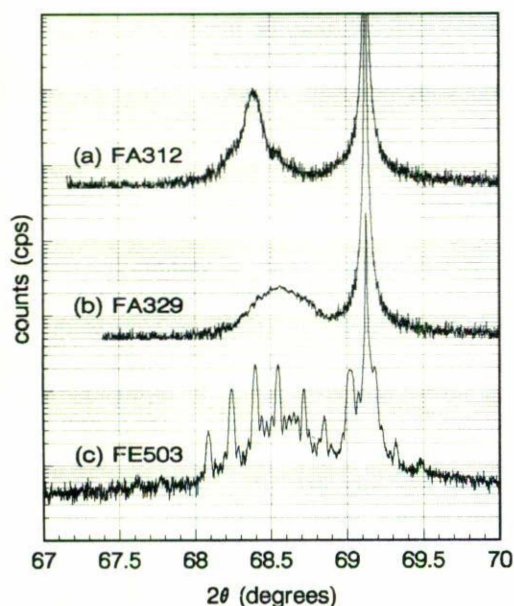

FIGURE 1. X-ray diffraction (400) rocking curves from (a) FA312-strained $Si_{0.89}Ge_{0.11}$/Si(001) alloy, the FWHM of SiGe peak is 180 arcsec, which indicates a high crystal quality of the $Si_{1-x}Ge_x$ alloy; (b) FA329-partially relaxed $Si_{0.89}Ge_{0.11}$/Si(001) alloy, the rocking curve of SiGe epilayers displays a very broad peak and the FWHM of SiGe peak is 680 arcsec, which primarily originates from misfit dislocations generated by the relaxation; and (c) FE503-$Si_{0.89}Ge_{0.11}$/Si(001) MQWs, besides the strong substrate and alloy reflections, Pendelossung fringes are seen on each side of the alloy peak, which shows good crystalline quality and abrupt interface of MQWs.

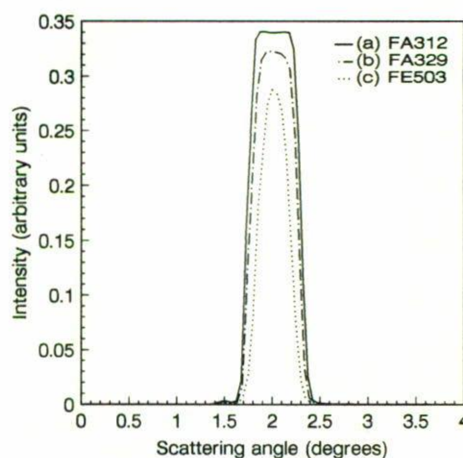

FIGURE 2. Measured angular-scattering distributions for $s \to s$ polarization combination from surfaces of (a) FA312-strained $Si_{0.89}Ge_{0.11}$/Si(001) alloy, the top of the scattering profile is nearly flat; (b) FA329-partially relaxed $Si_{0.89}Ge_{0.11}$/Si(001) alloy, the top of the scattering profile is nearly round; (c) FE503-$Si_{0.89}Ge_{0.11}$/Si(001) MQWs, the scattering profile displays Gaussian-like profile, which is different from those of FA312 and FA329.

composition in $Si_{1-x}Ge_x$ alloys as a function of the GeH_4 flow rate and the results of Auger electron spectroscopy (AES), corresponding thickness of $Si_{1-x}Ge_x$ epilayers is 350 nm. The rocking curve of SiGe epilayers exhibits a very broad peak, and the FWHM of SiGe peak is 680 arcsec, which primarily originates from misfit dislocations generated by the relaxation, because the $Si_{0.89}Ge_{0.11}$ epilayers exceeded the critical thickness and were partially relaxed [13, 14]. Figure 1c displays the rocking curve of $Si_{0.89}Ge_{0.11}$/Si MQWs (FE503). Besides the strong substrate and alloy reflections, Pendelossung fringes are seen on each side of the alloy peak. These oscillations are predicted by dynamical theory and result from interference between the incident wave field and the wave field reflected from the back surface of the crystal, which shows good crystalline quality and abrupt interface of MQWs. The strain distributions in $Si_{1-x}Ge_x$ epilayers of these three samples are different, which may affect their surfaces.

Representative experimental results obtained from the scatterometer with angle resolved scattering for the 3 surfaces of $Si_{1-x}Ge_x$/Si samples are plotted in Figs. 2 and 3, where we present measurements taken with $s \to s$ and $s \to p$ polarization combinations, respectively. For $s \to s$ polarization state (also $s \to p$ polarization combination) the scattering profiles are different for the samples with different strain distributions, even if all samples display mirror-like surface morphology. The top of the scattering profiles is nearly flat for FA312 and round for FA329. The scattering profiles of sample FE503, however, show Gaussian-like profile. The intensity and width of the profiles for FA312 are biggest. They are smallest for FE503. The sample FE503 with the narrowest width of $s \to s$ polarization state presents the lowest normalized intensity. However, its normalized intensity of $s \to p$ polarization state increases. The results mean that the surfaces of $Si_{1-x}Ge_x$/Si heterostructures are microrough to different degree for the samples with different strain distributions.

There is a 4.2% lattice mismatch between Si and Ge. However, because this is a strongly bonded system, the deposition of $Si_{1-x}Ge_x$ (or alternatively the co-deposition of $Si_{1-x}Ge_x$ and Si) onto a Si (100) surface leads to the formation of a strained epilayer. The $Si_{1-x}Ge_x$ epilayer adopts the in-plane lattice parameter of the Si (100) template, which results in a tetragonal distortion of the unit cell to accommodate the strain. Growth of strained layers continues until a "critical thickness" is reached, at which point it becomes favorable for the layer to relax. DCXRD (004) rocking curves for relaxed SiGe layers exhibit very broad peaks (Fig. 1b), in contrast to strained layers (Fig. 1a). From Figs. 2 and 3 the scattering profiles are different with samples FA329 and FA312, which shows the surface of sample FA329 is different from that of sample FA312. That is, the surface profile of partially relaxed $Si_{1-x}Ge_x$ is different from that of strained $Si_{1-x}Ge_x$. For MQWs sample FE503, $Si_{0.89}Ge_{0.11}$ and Si alternatively co-deposit onto a Si (100) surface, which leads to the formation of a strained $Si_{0.89}Ge_{0.11}$ and Si epilayers, i.e., the compressive strain within $Si_{0.89}Ge_{0.11}$ epilayers and tensile strain within Si layers. Obviously, the strain distribution

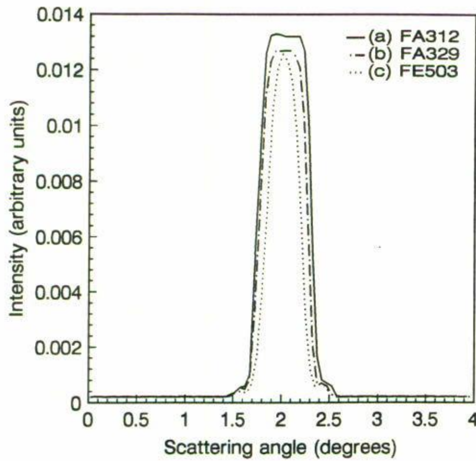

FIGURE 3. Measured angular-scattering distributions for $s \to p$ polarization combination from surfaces of (a) FA312-strained $Si_{0.89}Ge_{0.11}$/Si(001) alloy; (b) FA329-partially relaxed $Si_{0.89}Ge_{0.11}$/Si(001) alloy; (c) FE503-$Si_{0.89}Ge_{0.11}$/Si(001) MQWs. The increase of the normalized intensity of $s \to p$ polarization state for sample FE503 suggests that the interface of $Si_{0.89}Ge_{0.11}$/Si MQWs is more abrupt than that of $Si_{0.89}Ge_{0.11}$ alloys.

is different from that in $Si_{0.89}Ge_{0.11}$ epilayers of samples FA329 and FA312, which is confirmed by DCXRD (004) rocking curves (Fig. 1). According to its scattering profiles, Gaussian-like profile, which are different from those of FA312 and FA329 (Figs. 2 and 3), the surface of FE503 is different from that of samples FA329 and FA312. This implies that the surfaces of heterostructures are related to the strain distribution in epilayers. Note in Figs. 2 and 3 that the $s \to s$ polarization state of sample FE503 presents the lowest normalized intensity in these 3 samples, whereas its normalized intensity of $s \to p$ polarization state increases. Pendelossung fringes of X-ray diffraction (400) rocking curves from FE503 show good crystalline quality and abrupt interface between Si and $Si_{0.89}Ge_{0.11}$ epilayers (Fig. 1c). The increase of the normalized intensity of $s \to p$ polarization state for sample FE503 suggests that the interface of $Si_{0.89}Ge_{0.11}$/Si MQWs is more abrupt than that of $Si_{0.89}Ge_{0.11}$ alloys, which is consistent with the results of X-ray diffraction (400) rocking curves.

4. Summary

$Si_{1-x}Ge_x$ pseudomorphic layers at various germanium fractions, x, in the alloy were grown by gas source molecular beam epitaxy with Si_2H_6 and GeH_4 as sources on (100) Si substrates. The structural characterization of $Si_{1-x}Ge_x$ pseudomorphic layers was investigated by double-crystal X-ray diffractometry, and the surface characterization was studied using a scatterometer with angle-resolved scattering for the first time. The signals with a scattering angle of $\pm 2°$ around the specular direction for details of the polarization of light scattering were measured. Strained and partially relaxed $Si_{1-x}Ge_x$ alloys and $Si_{1-x}Ge_x$/Si multi-quantum wells (MQWs) present different scattering profiles, even if all of them display mirror-like surface morphology, which indicates that the surfaces of heterostructures are microrough to different degree with the samples grown under different growth conditions. The increase of the normalized intensity of $s \to p$ polarization state for $Si_{1-x}Ge_x$/Si MQWs suggests that the interface of $Si_{1-x}Ge_x$/Si MQWs is more abrupt than that of $Si_{1-x}Ge_x$ alloys. Experimental results show that surfaces have a closed connection with the strain distributions. We stress the importance of our results for the application of optical scattering technique as an optical probe for the study of surface microroughness in semiconductor heterostructures grown on lattice-mismatched surface.

Acknowledgment

Drs. L.-F. Zou, S.E. Acosta-Ortiz, and L.E. Regalado, Mr. J. Sarabia-Torres, and M. Sc. G.A. Perez-Herrera would like to acknowledge the support from Consejo Nacional de Ciencia y Tecnología (CONACyt)-México.

* Corresponding author, Tel. (52-4) 915-4325, Fax: (52-4) 918-3223. e-mail: lfzou@cioags.com.mx & lfzou@hotmail.com.

1. G.L. Patton, *IEEE Electron Device Letters* **11** (1990) 171.
2. T.L. Lin, *IEEE Trans. on Electron Devices* **38** (1991) 1141.
3. J.M. Baribeau, *Appl. Phys. Lett.* **52** (1988) 105.
4. J.C. Stover, *Optical Scattering: Measurement and Analysis*, (SPIE Optical Engineering Press, Washington, 1995).
5. T.A. Germer, C.C. Asmail, and B.W. Scheer, *Opt. Lett.* **22** (1997) 1284.
6. T.A. Germer and B.W. Scheer, *Proc. SPIE* **3426** (1998) 160.
7. L.F. Zou *et al.*, *J. Chin. Electron. Microsc. Soc.* **16** (1997) (in English) 395.
8. L.F. Zou *et al.*, *Rev. Mex. Fís.* **44** (1998) 93.
9. A. Ishizaka and Y. Shiraki, *J. Electrochem. Soc.* **85** (1986) 666.
10. L.F. Zou *et al.*, *Chin. J. Semiconductors* **18** (1997) 333.
11. L.D. Landau and E.M. Lifshitz, *Theory of Elasticity*, (Pergamon, New York, 1960).
12. R.W. James, *Solid State Phys.* **15** (1963) 169.
13. L.F. Zou *et al.*, *Appl. Phys. Lett.* **72** (1998) 845.
14. D.C. Houghton, *J. Appl. Phys.* **70** (1991) 2136.