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Interfacial Misfit Array Technique for GaSb Growth on GaAs (001) Substrate by Molecular Beam Epitaxy

D. BENYAHIA,[1,3] Ł. KUBISZYN,[2] K. MICHALCZEWSKI,[1] A. KĘBŁOWSKI,[2] P. MARTYNIUK,[1] J. PIOTROWSKI,[2] and A. ROGALSKI[1]

1.—Institute of Applied Physics, Military University of Technology, 2 Kaliskiego Str., 00-908 Warsaw, Poland. 2.—Vigo System S.A., 129/133 Poznańska Str., 05-850 Ożarów Mazowiecki, Poland. 3.—e-mail: djalal.benyahia@wat.edu.pl

Undoped GaSb epilayers, deposited at low growth temperature (440°C), have been grown on GaAs (001) substrate with 2° offcut towards [110], by a molecular beam epitaxy system. Interfacial misfit array (IMF) growth mode has been used in order to impede the propagation of the threading dislocations through the GaSb epilayer. Under optimized growth parameters, both transmission electron microscopy (TEM) measurements and high-resolution x-ray diffraction (HRXRD) revealed the presence of a periodic array of pure 90° edge dislocations along [110]. Furthermore, HRXRD shows a full width at half maximum of a 2-μm-thick GaSb epilayer peak as low as 195 arcsec. In addition, the GaSb layer is found to be 99.8% relaxed, with a residual strain of 1.4×10^{-4}. Moreover, based on TEM measurements, the dislocations spacing or the period of the IMF was found to be 5–5.2 nm.

Key words: Molecular beam epitaxy, GaSb, IMF, high-resolution x-ray diffraction, semiconductor III–V

INTRODUCTION

Antimonide-based compound semiconductors have peculiar applications in a broad range of optoelectronic and electronic systems, thanks to their outstanding band alignments, small effective mass, and high electron mobility.[1–3] Hence, considerable attention has been given to these materials and uge progress has been achieved in the devices' growth and invention; for instance, semiconductor lasers,[4] field effect transistors,[5] and infrared detectors.[6] Though recent developments have allowed the growth of high-quality lattice-matched GaSb epitaxy on native substrates, GaAs substrates are favored for many applications. This is due to their low cost, semi-insulating and suitable thermal properties, GaAs is transparent to more active regions, and it forms good n and p ohmic contact. However, the GaAs substrate and GaSb material exhibit a 7.8% lattice mismatch which results in a high density of threading dislocations, typically on the order of 10^9 cm^{-2}.[7] This latter is highly detrimental to the electrical and optical properties of the device structures. Several techniques have been used to mitigate this deleterious effect, including metamorphic buffer layers,[8] strain-relief superlattice,[9] and interfacial misfit dislocation (IMF) growth mode.[10] In the metamorphic buffer layer approach, the strain is accommodated through tetragonal distortion in addition to defect formation, within a critical thickness. This technique shows several shortcomings, such as the indispensability to grow thick buffer layers (often >1 μm), poor thermal and electrical conductivity, and crucial material degradation through the formation of threading dislocations. In IMF growth mode, the strain is relieved instantaneously at the interface between the GaAs substrate and the GaSb epilayers by the formation of (2D) periodic IMF arrays consisting of pure-edge 90° dislocations along both [110] and [1-10] directions.[11] The growth of thick GaSb epilayers on GaAs substrates was considered to begin as islands and then merge to create layers, and in this

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consideration, both 60° and 90° misfit dislocations exist.[12] However, the dominating strain relief mechanism was supposed to be 90° misfits, while the minority 60° ones were demonstrated to give birth to threading dislocations. The origin of the 60° misfits is still ambiguous, but it is believed to be related to three factors: island coalescence, the growth temperature, and the degree of the mismatch. Island coalescence has been demonstrated to cause 60° edge dislocations.[10] The growth temperature has been proved to be a robust factor in defining which misfit is formed: GaSb layers grown at ~520°C favor 90° misfits, while at ~560°C they favor 60° ones.[13,14] The lattice mismatch has been demonstrated to be a major factor in the formation of 90° misfits. Low strain (<2%) gives birth to 60° misfits, moderate strain (3–4%) results in mixed 60° and 90° misfits, and large strain (>6%) results in pure-edge 90° misfits.[15] There are some conflicts in IMF growth techniques across the literature. While all the recent groups confirm that the desorption of arsenic from the GaAs surface is the primary preoccupation, the option to switch from arsenic to antimony is performed at boththe GaAs growth temperature andthe GaSb growth temperature.[16] Transmission electron microscopy (TEM), etch pit density (EPD), and surface probe microscopy have been utilized to determine the threading dislocation density. However, these techniques have the disadvantage of being destructive methods. In addition, EPD measurements give an underestimation for dislocation densities greater than $\sim 5 \times 10^6$ cm^{-2}.[17] Recently, non-destructive and large-area measurements have been performed by using x-ray diffraction (XRD) to analyze the threading dislocations formation.[18–20] These methods also have other advantages: the quantitative investigation such as the strain and the relaxation of GaSb epilayer on the GaAs substrate.

In this work, we report on the possibility to grow GaSb epilayers with the IMF approach on a GaAs (001) substrate at low growth temperature (440°C) in comparison to that reported by other groups (510°C).[10] The surface morphology has been assessed by Nomarski optical microscopy and high-resolution optical profilometry. In addition, cross-sectional TEM has been used to investigate the heterointerface GaAs/GaSb. The strain relaxation, dislocations density, and IMF properties have been characterized by high-resolution x-ray diffraction (HRXRD).

EXPERIMENTAL

The samples have been grown on GaAs (001) substrates with 2° offcut towards [110] in a RIBER COMPACT 21-DZ solid-source molecular beam epitaxy system, equipped with valved crackers for As and Sb sources, and a BandiT system to monitor the substrate temperature. After thermal desorption of the oxide at 600°C under As_4 overpressure, a 25-nm-thick GaAs buffer layer was deposited at 580°C in order to get a smooth starting surface. The reflection high-energy electron diffraction (RHEED) pattern exhibits a 2×4 (Fig. 1a) reconstruction indicating a flat As-rich surface.

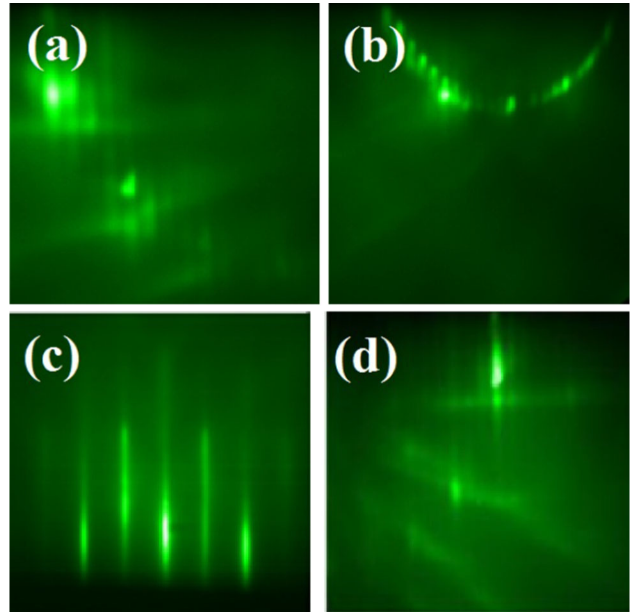

Fig. 1. GaSb layer RHEED patterns (a) (×4) of the GaAs surface, (b) (×8) of the Sb-soaked Ga-rich surface, (c) (×1) of the GaSb surface (d) and (×3) of the GaSb surface.

For the growth of GaSb layers, after the GaAs deposition, the As and Ga shutters were closed to let the As adatoms desorb from the surface in order to have a Ga-rich surface. Theoretically, the RHEED pattern should change from(2 × 4 to 4 × 2. In our growth, the 2 × 4 pattern becomes faint, while the ×2 reconstruction transforms almost to a ×3 one. This Ga-rich surface is considered as the key for constituting highly periodic misfits. After waiting for about 30 min for the appearance of 4 × 2, the substrate temperature is cooled to 440°C under Sb overpressure. Interestingly, the RHEED pattern transforms to a 2 × 8 reconstruction (Fig. 1b) revealing that the Sb adatoms have incorporated and the equilibrium state has been reached.[21] This reconstruction is proof of atomic packing instead of tetragonal distortion. Once the substrate temperature is stabilized at 440°C, the Ga beam flux is introduced, and the GaSb layer growth is initiated. Immediately, the RHEED exhibits a spotty pattern for the first few monolayers indicating a three-dimensional growth mode. During the growth of the next few monolayers, the RHEED shows a streaky 1 × 3 pattern (Fig. 1c, d), which is evidence of a two-dimensional growth mode. Consequently, the GaSb growth is a Volmer–Weber growth mode. The growth rate for the GaSb layer was 0.76 μm/h while the BEP (beam equivalent pressure) group V/III flux ratio (Sb/Ga) was set to be 5.

In order to protect the surface, the samples were cooled under Sb overpressure until 300°C, meanwhile the RHEED exhibits a 1 × 3 pattern.

The structural quality and the defect properties of the grown samples were assessed by cross-sectional TEM. These measurements have been carried out on a FEI Talos microscope operating at 200 kV. Cross-sectional TEM specimens were got ready by a mechanical pre-thinning followed by an Ar ion milling. HRXRD of PANalytical X'Pert was used to investigate the crystallographic properties and the dislocation density in the GaSb layers. Furthermore, it was used to characterize the IMF array. The Cu Kα_1 radiation ($\lambda \approx 1.5406$ Å) originating from a line focus was used. The x-ray beam was monochromatized by a four-bounce Ge (004) hybrid monochromator. The measurements were performed in both the ω and 2θ-ω directions. The in-plane measurements were performed in the [110] direction. On the other hand, the symmetric (004) and asymmetric (2-2-4) reciprocal space map (RSM) were performed. Surface morphology was assessed by Nomarski optical microscopy and high-resolution optical profilometry.

RESULTS AND DISCUSSION

Figure 2 shows the surface morphology of undoped GaSb layers assessed by Nomarski optical microscopy. As can be seen, the surface of the grown sample was shiny mirror-like, under optimized growth conditions of growth temperature of 440°C and a Sb/Ga flux ratio of 5. A surface roughness as low as 3.3 nm was obtained under these conditions. On the other hand, the same surface quality was obtained under a group V/III flux ratio of 5 and at 250–540°C temperature range.[22] Rough surfaces (Rq = 145.2 nm), as can be noticed in Fig. 2b, were collected under a growth temperature of 420°C and a Sb/Ga flux ratio of 3.3.

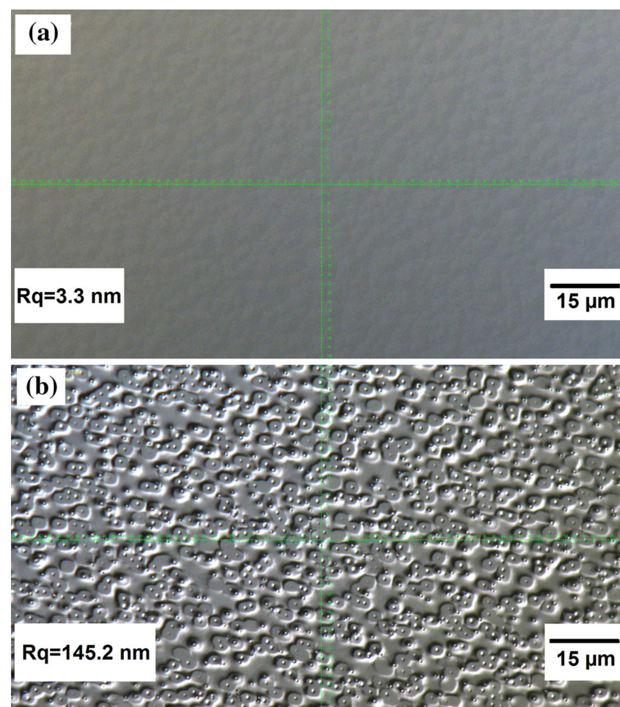

Fig. 2. The Nomarski optical microscopy pictures with a magnification of ×1000 of a 2-μm-thick GaSb layer grown (a) under optimized parameters and (b) under unoptimized parameters.

The investigation of the interfacial misfit array using cross-sectional TEM image is shown in Fig. 3. This clearly shows the presence of a periodic array of misfit dislocations along [110]. The periodicity of the IMF array is measured to be 5–5.2 nm. The strain generated by the large lattice mismatch is relieved spontaneously by the formation of the IMF array at the interface. Analogous misfit dislocation periodicity was reported in GaSb growth on a GaAs substrate by Ref. 10. References 7 and 12 who proposed that the quasi-perfect relaxation of GaSb is effectively viable due to the high efficiency of the 90° misfits in the strain mismatch relieving.

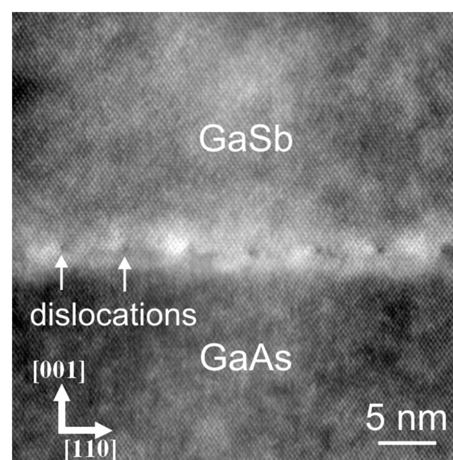

Fig. 3. Cross-sectional TEM imaging of GaSb epilayers grown on a GaAs substrate.

The 2θ-ω scan of symmetric (004) and asymmetric (2-2-4) HRXRD spectra for a 2-μm-thick GaSb layer grown using IMF technique are shown in Fig. 4a and b, respectively. The position of the GaSb layers depends on the reflection and the strain between the layer and the GaAs substrate. The peak separation between the GaAs substrate andthe GaSb layers could be solely determined to be 9590 arcsec, which corresponds to the theoretical value of a relaxed GaSb layer (9554 arcsec). During the cooling of the substrate temperature, after the growth of the GaSb layer, the cubic structure can be distorted to a tetragonal one. The perpendicular lattice parameter "c", and the parallel one "a", to the GaAs/GaSb interface, have been calculated from the 2θ-ω scans for symmetric (004) and asymmetric (2-2-4) reflections, respectively (Fig. 4), using the followings equations:

$$c = 2\lambda/\sin(\theta_{004}) \quad (1)$$

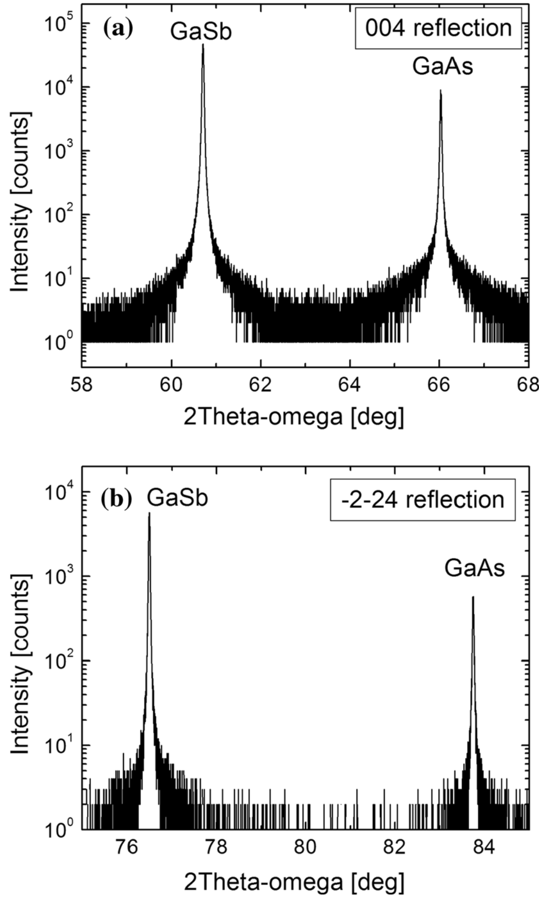

Fig. 4. HRXRD 2θ-ω scans for the GaSb layer grown on a GaAs substrate for (a) symmetric (004) reflection and (b) asymmetric (2-2-4) reflection.

$$\frac{1}{d_{hkl}^2} = \frac{h^2 + k^2}{a^2} + \frac{l^2}{c^2} \tag{2}$$

where λ is the wavelength of the x-ray beam, d_{hkl} is the spacing between the planes (hkl), here (hkl) = (2-2-4). While the relaxed lattice parameter has been calculated from the following equation:

$$a_{\text{relax}} = (c + 2ba)/(1 + 2b) \tag{3}$$

where $b = (1 - \nu)/(1 + \nu)$, and ν is the Poisson ratio determined by $\nu = C_{12}/(C_{11} + C_{12})$, where the elastic stiffness parameters C_{11} = 8.85 × 10^{10} N/m^2 and C_{12} = 4.04 × 10^{10} N/m^2.[23]

The in-plane lattice parameter "a" is determined to be 6.0942 Å, while the perpendicular one is 6.0959 Å. On the other hand, the relaxed lattice parameter is 6.0950 Å. The relaxation, which is defined by Eq. 4, is found to be 99.8%.

$$R = (a - a_{\text{subst}})/(a_{\text{relax}} - a_{\text{subst}}) \tag{4}$$

GaSb layers grown on a GaAs substrate undergo biaxial compressive strain because of the lattice mismatch, and a tensile strain due to the difference in thermal expansion coefficient between GaSb (7.75 × 10^{-6} K^{-1}) and GaAs (5.7 × 10^{-6} K^{-1}).[24,25] Generally, the strain generated by the lattice mismatch is quickly relieved through the formation of misfit dislocations. The residual strain is the consequence of the difference between the thermal expansion coefficients of the epilayer and the substrate. Vertical strain ε in the GaSb layers is determined to be 1.4 × 10^{-4} by the following equation:

$$\varepsilon = (c - a_{\text{relax}})/a_{\text{relax}} \tag{5}$$

The dislocations in the single-crystal semiconductor causes the broadening of the rocking curve in two ways: (1) it introduces a rotation of the crystal lattice, which immediately broadens the rocking curve; and (2) these dislocations are encircled by a strain field, in which the Bragg angle of the crystal is not uniform.[18] The dislocations density (D) can be calculated from rotational broadening (Eq. 6) and from the strain broadening (Eq. 7) as follows[18]:

$$D = K_\alpha/4.36b^2 \tag{6}$$

$$D = K_\varepsilon/0.090b^2 \left|\ln\left(2 \times 10^{-7}\text{cm}\sqrt{D}\right)\right| \tag{7}$$

where b is the Burger's vector, in our case, $b = a/2$.[10] While K_α and K_ε are two parameters determined from the following equation:

$$\beta^2 = K_\alpha + K_\varepsilon \tan^2\theta \tag{8}$$

where β is the measured full width at half maximum (FWHM) of the GaSb epilayer. Four rocking curves were performed at different values of θ (Table I). Having plotted β^2 as a function of $\tan^2\theta$, the parameters K_α and K_ε are determined from the intercept and the slope (Fig. 5), respectively. One can readily calculate K_α and K_ε to be 32,568 arcsec^2 and 5184 arcsec^2, respectively. Thus, the dislocations density determined from the rotational broadening is 1.89 × 10^8 cm^{-2}, while based on the strain broadening, D is equal to 2.53 × 10^8 cm^{-2}, which is in suitable agreement with the previous result. This agreement confirms the presence of 90° misfit dislocations.[18] In the existence of 60° misfit dislocations, the threading dislocation density determined by the strain broadening is reduced by a factor of two compared to that calculated from the rotational broadening. Reference 10 have reported a dislocation density of ~10^5 cm^{-2}.

The dislocation type can be identified by two parameters: the angle ϕ of the 2-2-4 RSM and the FWHM ratio for both the symmetric and rocking curves of the 004 reciprocal lattice point (RLP).[26] Figure 6 exhibits the RSM of the GaSb layers for the 004 and 2-2-4 reflections. The first RSM is used to calculate the GaSb 004 FWHM ratio between Q_z and Q_x directions, while the second one is required to determine the angle ϕ between the FWHM and the

Table 1. X-ray rocking curve data for a 2-μm-thick GaSb layer grown on a GaAs substrate using the IMF technique

(hkl)	θ, °	Tang$^2\theta$	β, arcsec
(004)	30.38	0.34	195
(115)	41.02	0.76	200
(117)	64.47	4.38	238
(2-2-4)	38.24	0.79	192

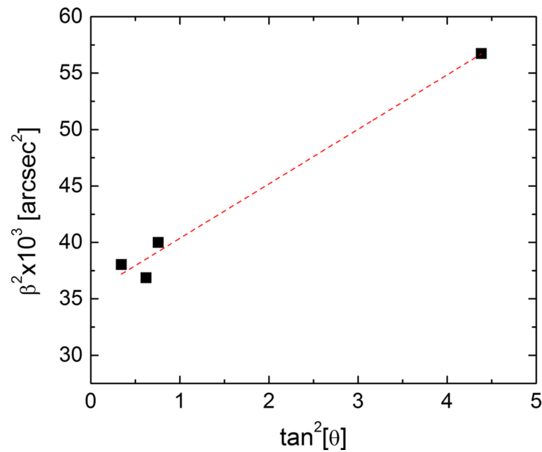

Fig. 5. The β^2 versus tan$^2\theta$ for a 2-μm-thick GaSb layer grown on a GaAs substrate.

Q_x direction. The expected value of ϕ can approach 38° for the 90° misfits dislocation, and 36° in the presence of 60° dislocations. The 2-2-4 RSM plotted in Fig. 6b revealed a value of 41.0° for the angle ϕ. On the other hand, the FWHM ratio is believed to be 0.37 for 60° edge dislocations and 0.71 for the 90° ones. From Fig. 6a, this ratio is found to be 0.30. Reference 20 demonstrates that both parameters ϕ and FWHM ratio are thickness-dependent. For instance, the angle ϕ shifts from 33° to 37° (FWHM ratio increase from 0.4 to 0.55), when the GaSb thickness increases from 0.25 μm to 1.5 μm. The precedent utilization of this method (especially GaAs on Si) were in the case of a thick buffer layer which were more probably completely relaxed.[27] The superelliptical shape of the asymmetric RSM shown in Fig. 6b gives ample proof that the IMF array of the 90° edge dislocations is formed.[26]

Reference 26 report on a simple method to characterize a correlated dislocations array. The relationship between misfit dislocations is as follow:

$$\Delta Q = K\sqrt{\gamma\rho/d} \tag{9}$$

where ΔQ is the scattering vector, d is the thickness of the GaSb layer, ρ is the dislocation line density, γ is the correlation factor, and K is a parameter determined by the scan direction in reciprocal

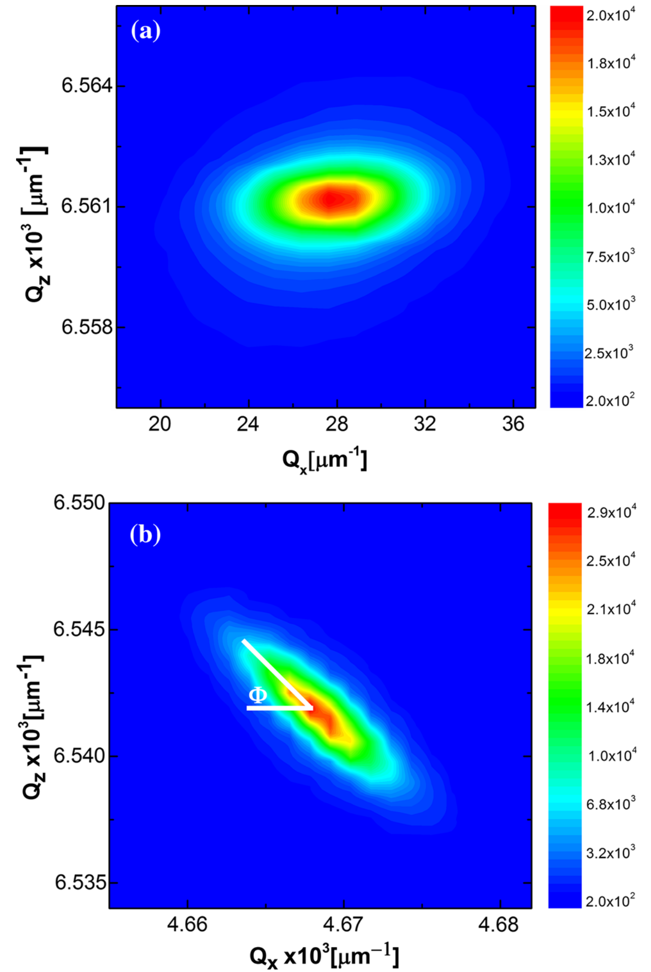

Fig. 6. RSM of GaSb layers grown on a GaAs substrate for (a) symmetric 004 reflection and (b) asymmetric 2-2-4 reflection.

space, the dislocation type, and the RLP.[26,28] For GaSb layers grown on the GaAs substrate, the K parameter is 11.805 for ΔQ_x and 8.347 for ΔQ_z.[29] Moreover, the correlation factor γ is equal to 0.015 for ΔQ_x and 0.005 for ΔQ_z.[20] Thus, the dislocations line density is 0.1495 nm^{-1}, which leads to a dislocations spacing of 6.7 nm. This is greater than that measured by TEM (which is the accurate value), and we believe that this difference is within the experiment error.

SUMMARY AND CONCLUSIONS

In summary, high-quality GaSb epilayers have been grown on a GaAs (001) substrate using Ithe MF approach. This growth was performed at low growth temperature compared to that reported in the literature. A mirror-like surface was obtained with a roughness of 3 nm. The presence of Ithe MF array of 90° pure-edge dislocations along [110] was confirmed by TEM measurements with a periodic spacing of 5–5.2 nm. On the other hand, the IMF presence is also confirmed by HRXRD analysis. The GaSb layers have been found to be 99.8% relaxed, with a residual strain of $\sim 10^{-4}$. The dislocations density was determined to be as low as 10^{-8} cm^{-2}. Future work should look to decreasing dislocations density.

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