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A strain relief mode at interface of GaSb/GaAs grown by metalorganic chemical vapor deposition

W. Zhou,[1,a)] W. Tang,[1] and K. M. Lau[2]
[1]*State key Laboratory of Electronic Thin Films and Integrated Devices, University of electronic Science and Technology of China, No. 4 the Second Block, North Road, Chengdu City 610054, China*
[2]*Department of Electronic and Computer Engineering, Photonics Technology Center, Hong Kong University of Science and Technology, Clear Water Bay, Kowloon 0085, Hong Kong*

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An "atomic chain-like" array distinct from interfacial misfit dislocation arrays was characterized at the interface of GaSb/GaAs grown by metalorganic chemical vapor deposition. Using high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) and x-ray energy-dispersive spectroscopy, we obtained the chemical composition of this structure and confirmed significant anion intermixing within the GaAsSb alloy layer at the interface. Atomic-scale HAADF-STEM imaging and geometric phase analysis revealed variations in the local strain field, indicating that the GaAsSb layer can partially relax the lattice misfit strain. Our results indicate that self-organized alloy intermixing during GaSb epilayer growth on GaAs provides strain relief. © *2011 American Institute of Physics*. [doi:10.1063/1.3663571]

Since strain-relaxed epilayers have emerged as templates for high-performance transistors,[1–3] the development of several epitaxial growth techniques for high-quality thin films has attracted much attention. Recently, metamorphic epitaxy of larger lattice misfit Sb-based materials on GaAs has shown promise for applications in electronic and optoelectronic devices owing to their unique band-structure alignments, small electron effective mass, and high electron mobility.[4–6] Previous reports examining metamorphic epitaxy of GaSb on GaAs have focused on the relaxation of misfit strain by dislocations.[7–9] However, alloy intermixing and lateral composition modulation are anticipated at the GaAs/GaSb interface,[10,11] and they are believed to relax strain through surface-enhanced roughening.[12] Thus, we considered the effect of alloy intermixing on strain relaxation in the growth of a GaSb epilayer on GaAs.

We show that a strain-relaxing buffer layer of unusual thickness exists between the GaSb epilayer and the GaAs substrate. The 2D growth process significantly differs from those previously reported.[7–9] The GaSb epilayers were grown on semi-insulating GaAs (001) substrates by low-pressure metalorganic chemical vapor deposition (MOCVD). To study the effect of intermixing on strain relaxation, samples were prepared using interfacial misfit (IMF) and non-IMF growth conditions, producing samples without and with the intermixing buffer layer, respectively. In IMF growth mode, AsH_3 flux was stopped and the surface was exposed to H_2 flux for 30s to remove excess As at the surface of GaAs buffer layer; for non-IMF growth condition, AsH_3 flux was stopped at the surface of GaAs buffer following the growth of GaSb and the V/III ratio is varied from 1.4 to 1.25. The more details have been reported elsewhere.[10]

Although useful for studying crystal structure, transmission electron microscopy (TEM) cannot routinely determine local lattice parameters qualitatively. However, the combination of scanning tunneling microscopy (STM) and high-angle annular dark-field (HAADF) imaging can provide chemical information with atomic resolution comparable to that of TEM.[11] HAADF scanning transmission electron microscopy (HAADF-STEM) was performed using a Tecnai 20 D522 S-Twin field-emission electron microscope, equipped with HAADF detector. In addition, the cross-sectional x-ray energy-dispersive spectroscopy (XEDS) line profile was recorded at 50 μA in an aberration-corrected 200-kV instrument with a lithium drifted silicon [Si(Li)] detectors, while the experimental integration time was 1 min.

Bright-field cross-sectional TEM images of GaSb/GaAs films fabricated under IMF and non-IMF growth conditions are shown in Figs. 1(a) and 1(b), respectively. Fig. 1(a) shows a clear IMF dislocation array at the GaSb/GaAs interface along the [1-10] direction, in agreement with recent reports.[7–9] Additionally, we found an elongated column that we term "atomic chain-like" along the [001] direction with a periodic array along the [1-10] direction. Notably, no threading dislocations or dark-line defects are detectable in the GaSb epilayer in either image. As shown in Fig. 1(b), there are two length scales of atomic chain-like columns arranged alternately along [1-10] with a periodicity of 5.56 nm, which is nearly equal to that of the IMF array.[7–9] We attribute this to the same strain relaxation by misfit dislocation. However, the atomic chain-like column array is $\sim$30 nm in length along the growth direction, an order of magnitude larger than that of the IMF array ($\sim$3 nm).

For atomic resolution chemical information, a low magnification HAADF-STEM image of a sample prepared by non-IMF growth mode was acquired (Fig. 2(a)). As seen in Fig. 1(b), there are two length scales of atomic chain-like columns arranged alternately along [1-10] in the GaSb layer, which may be formed by lattice strain relaxation by misfit dislocation and localized composition fluctuations resulting from anion intermixing at the GaSb/GaAs interface.[12] Compared with the IMF array, this sample fabricated by non-IMF mode has reduced strain energy and a modified growth mode

a)Author to whom correspondence should be addressed. Electronic mail: weizhou@uestc.edu.cn.

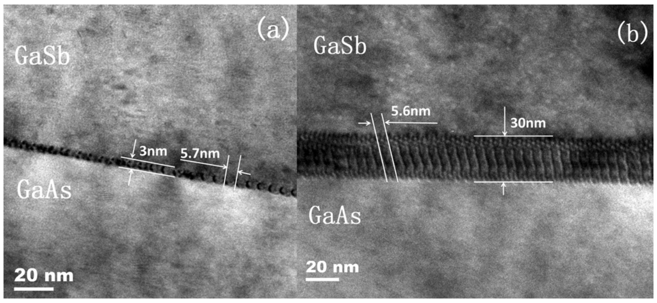

FIG. 1. Cross-sectional bright-field TEM images of the GaSb layer on GaAs (a) grown by IMF growth mode and (b) grown by non-IMF growth mode. The dot-like structures are the damaged areas created by ion milling.

may cause by the intermixing buffer layer, the difference can be best understood by considering the interfacial anion mixing due to the significant Sb segregation at low V/III ratio (1.25) and excess As adatomic at the surface of GaAs.

To demonstrate anion intermixing at the GaSb/GaAs interface, a higher magnification HAADF-STEM image with corresponding XEDS analysis was acquired (Fig. 2(b)). The dark line marks the location of the XEDS analysis and the arrow indicates the GaSb/GaAs interface. XEDS was performed across the modulated regions along [1-10] in order to calculate the amplitude and periodicity of the composition modulation of each element. The integrated signals are plotted as a function of position across the dark line for Ga, As, Sb, and HAADF intensity (Fig. 2(c)). XEDS results indicate that As and Sb atoms are both present ~4 nm from the interface and that the As and Sb contents oscillate in phase with each other. This implies intermixing of As and Sb and provides evidence that the broadened interface is a GaAsSb alloy buffer layer. The periodicity of the composition modulation of elements is ~5.6 nm, consistent with the periodicity of the atomic chain-like array as measured from TEM images. The composition of As and Sb atoms in the GaAsSb buffer layer can be estimated from XEDS according to the Cliff-Lorimer equation.[13] For the GaAsSb alloy layer, we can calculate the composition ratio of Sb and As related to Ga from

$$\frac{x_{Sb}}{x_{Ga}} = k_{SbGa}\frac{I_{Sb}}{I_{Ga}}, \qquad \frac{x_{As}}{x_{Ga}} = k_{AsGa}\frac{I_{As}}{I_{Ga}}, \tag{1}$$

where I_{Sb}, I_{As}, and I_{Ga} are the integrated intensities from Sb, As, and Ga, respectively. k_{SbGa} and k_{AsGa} are Cliff-Lorimer factors for the element pairs GaSb and GaAs, and they can be calculated from first principles or determined experimentally using standards of known composition. x_{Sb} and x_{Ga} are the mole fractions of Sb and Ga, respectively, at a given location. The k-factor is a sensitivity factor and is not constant. To calculate the values of x_{Sb} and x_{As} in a GaAsSb alloy, we used regions of the GaAs substrate and GaSb layer distant from the interface to determine the factors k_{SbGa} and k_{AsGa}. Fig. 2(d) shows that the values of x_{Sb} and x_{As} are dependent on position in the GaAsSb buffer layer as derived from Fig. 2(c) using the Cliff-Lorimer equation, where we assumed that the composition of Ga was an appropriate unity. The Sb and As mole fractions vary from 0.5 to 0.7 and from 0.34 to 0.48 along [1-10], respectively. The composi-

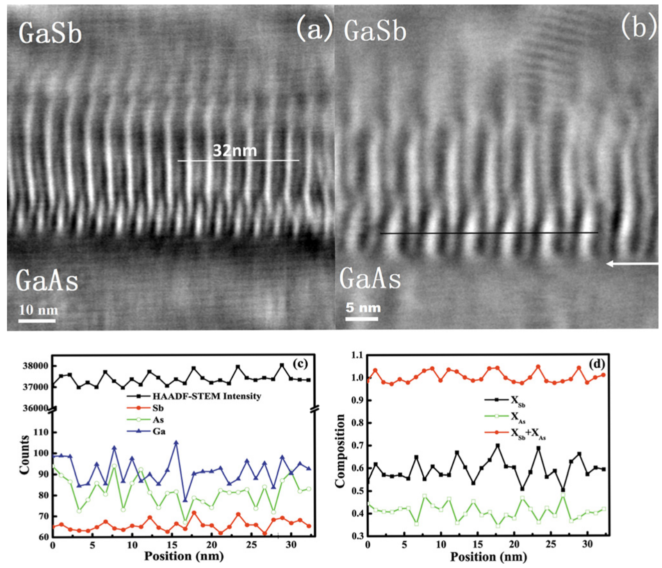

FIG. 2. (Color online) HAADF-STEM image of the interface from sample grown by non-IMF mode (a) at low magnification and (b) at higher magnification. The dark line marks the position of XEDS analysis and the arrow indicates the position of the interface of GaSb and GaAs. (c) Intensity profiles of Ga (solid triangles), As (solid circles), Sb (open circles), and HAADF intensity (solid squares). (d) The values of x_{Sb} and x_{As} depend on position in the GaAsSb buffer layer.

tion modulation may result from the lattice misfit strain of GaSb grown on GaAs.[12] The mean value (1.002 ± 0.0245) of sum of Sb and As mole fractions is nearly equal to unity (the theoretical value), and the small deviation probably arises from the assumption of Ga composition as unity; there are excess Sb atoms present in interstitial sites and Ga sites in the GaSb layer.[14] Even so, the chemical composition obtained using our method correctly indicates the lateral composition modulation at the GaSb/GaAs interface despite this minor deviation from the theoretical value.

As mentioned above, the alloy intermixing in strained epitaxy will affect the mode of strain relaxation by dislocation. Fig. 3(a) shows an atomic-scale high resolution HAADF-STEM image of the GaAsSb buffer layer. Wiener filtering was used to remove the noise and influences of probe. There is a high-contrast band along the growth direction ~10 nm wide on the cross-section (110) plane, which includes a bright band and two dark bands. Strain-induced HAADF contrast is formed by two well-known mechanisms. Dislocations in close proximity to one another cause strain that increases HAADF intensity; likewise, random strain at an atomic level produces reduced HAADF intensity.[15] The bright and dark contrast bands in Fig. 3(a) may be understood by these mechanisms; specifically, the central bright band likely contains a large number of dislocation (which would effectively relax the large lattice misfit strain) and the dark bands probably contain fewer point defects induced by a random strain field and composition anomalies.[16]

Fig. 3(b) shows that the ε_{xx} component of the strain field is parallel with the interface ([001] growth direction) as derived from Fig. 3(a) using the geometric phase analysis (GPA) method. The strain mapping image shows the larger lattice constant (positive apparent strain) of the interface defined with respect to the GaAs substrate reference lattice.

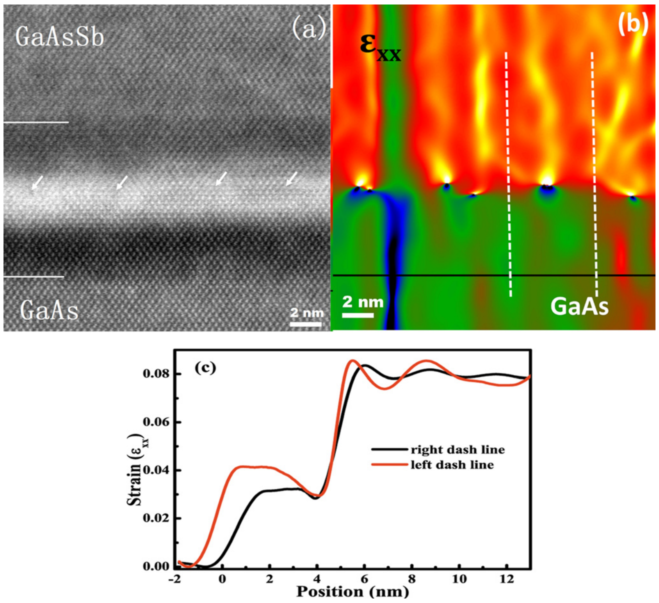

FIG. 3. (Color online) (a) Atomic-scale high resolution HAADF-STEM image from sample grown by non-IMF mode. The white lines indicate the interface of high-contrast regions and the white arrows mark the position of dislocations. (b) Strain map parallel (ε_{xx}) with the interface. The brighter dots indicate the dislocation core. (c) The local measurement of strain as a function of the distance perpendicular to the interface.

The dislocation cores correspond to the maximum strain field and are easily delineated at the nanometer scale (Fig. 3(b)); moreover, they appear in the bright band, confirming our conclusion that the bright zone contains a higher density of dislocations. The local measurement of strain as a function of distance perpendicular to the interface is plotted in Fig. 3(c). The calculated strain value from the line profile can be used to obtain the average strain field, which can then be related to the relaxation state of the layers in the growth direction. To avoid the effect of dislocation cores, two line profiles were taken in the middle of two dislocation cores, as shown in Fig. 3(b). Notably, at a position distant from GaAs, the strain increased to an initial value of 4% for the left line profile and 3% for the right line profile, and this was followed by a rapid increase to 7.8% at the position parallel to the dislocation cores. This result is in stark disagreement with previous reports regarding the relaxation of lattice misfit strain of a GaSb layer grown on GaAs,[7–9] where the strain state occurred only near the dislocation core and the large lattice misfit strain was completely relaxed to its bulk value within three unit cells to a few nanometers. In our experiment, excess atomic As coupled with Sb segregation led to the generation of a GaAsSb alloy layer at the interface. The lattice misfit strain of GaSb and GaAs was reduced due to the presence of the GaAsSb alloy layer,[17] and hence, a high strain state was observed before the misfit dislocation emerged. This growth mode indicates that there are two distinct regions of strain relaxation, the first region of strain relaxation is attributed to the presence of an As-into-Sb exchange at the GaSb-on-GaAs interface; the second region of strain relaxation dominates by misfit dislocation. Fig. 3(c) indicates that at locations distant from the interface, the apparent strain reaches the value of 7.8%, which is consistent with the lattice misfit between GaAs and GaSb and implies that the completely relaxed GaSb film with low defects can be grown on GaAs substrate by this growth mode except for IMF mode.

In conclusion, we discovered an orderly atomic chain-like array that is distinct from an IMF array on a GaSb layer. The unique structure was investigated by HAADF-STEM. Using XEDS technology, we quantitatively analyzed the chemical composition of the atomic chain-like array and confirmed significant anion intermixing that allows the fabrication of a GaAsSb alloy buffer layer between the GaSb epilayer and GaAs substrate. Finally, our local measurements of strain clearly demonstrate that the presence of the GaAsSb alloy layer can partially relax the large lattice misfit in the GaSb/GaAs system. These results demonstrate that there is a strain relief mode in the growth of GaSb films on GaAs by MOCVD under particular growth conditions.

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