Assessment of Structural Properties of InAs/GaSb Superlattice by Double Crystal X-Ray Diffraction and Cross-Sectional Scanning Tunnelling Microscopy

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Abstract. An investigation of the structural properties of InAs / GaSb superlattices was carried out by double crystal X-ray diffraction analysis and cross-sectional scanning tunnelling microscopy. It was demonstrated that the combination of these methods make it possible to describe in sufficient details the thickness and composition of the alternating layers as well as characterize the interfaces between them. The effect of MBE growth conditions (Sb flux) on the cation and anion segregation profiles and interface bond formation was studied. It was suggested that the use of high Sb flux during the GaSb growth helps to minimize As-for-Sb exchange at the InAs-on-GaSb heterojunction and preserve In-Sb type bonds.

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1 Introduction

There is a significant interest in heterostructures based on InAs/GaSb material system due to their unique type II broken gap alignment. This alignment gives rise to interesting and unusual electron properties, such as hybridization of the conduction InAs and valence GaSb band states due to coupling across the interface and spatial separation of electrons and holes [1-3]. A number of optoelectronic devices, such as infrared detectors and lasers, resonant interband tunnelling diodes, and high electron mobility transistors, have been produced based on these special properties [4-6]. Since InAs/GaSb heterojunction has no common atoms, two types of interfaces can be formed: one with predominantly Ga-As and one with predominantly In-Sb type of bonds. Due to the large difference between the lattice constants,
band gaps, and other physical properties of InSb and GaAs, electronic and optical properties of InAs/GaSb devices will be strongly effected by the interface bonding [7-9]. During molecular beam epitaxial (MBE) growth a significant Sb fraction is incorporated into InAs layers due to the anion segregation at InAs-on-GaSb interface and residual Sb background, while In migrates to GaSb layers due to the cation segregation at the GaSb-on-InAs interface. These phenomena prevent formation of compositionally abrupt interfaces and result into spatially varying composition profiles, making the device-quality control over interfacial structure and composition with these materials especially challenging. Therefore, a fast and reliable technique is necessary to study and characterize InAs/GaSb superlattices.

Transmission electron microscopy (TEM) combined with high-resolution X-ray diffraction (XRD) have been previously widely used to study thickness and composition of III-V and II-VI superlattices [10-13]. However, due to the cross incorporation and segregation effects, it is challenging to get a complete picture of the thickness and composition of individual layers in thin InAs/GaSb superlattices using these two techniques only.

In this paper we present the results of double crystal X-ray diffraction (DCXRD) analysis of InAs/GaSb superlattices. Results of XRD analysis were supplemented with the In and Sb segregation profiles that were obtained using cross-sectional scanning tunnelling microscopy (STM). XRD profiles were analyzed with a dynamic X-ray simulation program and a good correlation between experimental and simulated spectra was obtained. The effect of MBE growth conditions (Sb flux) on the interface bond formation was also studied. It was determined that the use of high Sb flux during the GaSb growth helps to minimize As-for-Sb exchange at the normal interface and to preserve In-Sb type bonds.

2 Experimental details

Three InAs/GaSb superlattice samples (SL 1, SL 2, and SL 3) were grown on Te-doped p-type (001) GaSb substrates in a VG-80 molecular beam epitaxy (MBE) system. This system is equipped with In, Ga, and Be solid sources, a valved As cracker and an unvalved Sb cracker sources (to provide As$_2$ and Sb$_2$, respectively), a reflective high-energy diffraction (RHEED) system, and a quadrupole mass-spectrometer. The growth was performed at around 475$^\circ$C, approximately 10$^\circ$C above the (1 × 3) to (1 × 5) surface reconstruction transition, observed by RHEED on the static GaSb surface. The growth rate was 0.5 monolayer (ML)/s both for InAs and GaSb.

The growth conditions were identical for all three samples studied with the only difference in the Sb flux used during the growth. The ratio of incident-to-consumed Sb$_2$ was measured by quadrupole mass-spectrometer and maintained at 1.6:1, 4.5:1, and 8.5:1 levels for SLs 1, 2, and 3, respectively.
The samples consisted of a 110 nm-thick GaSb:Be buffer layer followed by 20 14-ML InAs/28-ML GaSb repeats and the whole structures were covered by a 100 nm-thick GaSb cap layer. Each InAs-on-GaSb heterojunction (normal interface) was formed by a 3 second total growth interruption to promote InSb bonds. Each GaSb-on-InAs heterojunction (inverted interface) was formed by a 12 second Sb soak to promote Sb-for-As exchange and the formation of InSb interface bonds.

DCXRD measurements were performed along (004) reflection using Cu Kα1 radiation. The periodicity of the superlattice was estimated from the spacing between the satellite peaks and the average lattice constant was extracted from the position of the 0th order peak of the superlattice with respect to the peak of the GaSb substrate. The data was analyzed using a dynamical X-ray simulation program [14,15].

Cross-sectional STM was performed after the growth in a separate system. The buried interfaces were exposed by cleavage along a (110) plane in a separate ultra high vacuum chamber and examined with Pt-Ir tip. Negative bias was used to image the anion sublattice while positive bias was used to image the cation sublattice.

3 Results and discussion

The (004) reflection X-ray rocking curves for three superlattice structures studied are shown in Figures 1(a), 1(b), and 1(c). The dominant peak at the center of the figures represent the reflection from the GaSb substrate, and a weaker peak, to the right from it, is the 0th order peak. The lattice mismatch is negative for all three structures and varies from -0.20% for SL 1 to -0.11% for SL 3, indicating the InSb bond formation at the interfaces and Sb incorporation into InAs layers [16].

More than three satellite peaks on each side of the zero order peaks are visible in the DCXRD rocking curves, indicative of high quality periodic structures. The intensity of the satellite peaks on the higher angle side is weaker than these on the lower angle side, which is consistent with the previously reported results [17].

The observed satellite peaks positions are plotted in the form of $\sin \theta$ in Figure 2. The linear relation to the order of the peaks gives evidence that the observed peaks are diffraction peaks from the superlattice structure. From the slopes, the superlattice periods are calculated using the equation:

$$\frac{\lambda}{2L} = m \quad (A.1)$$

where $L$ is the superlattice period, $\lambda$ is the CuKα1 wavelength (1.5406 Å), and $m$ is the slope of the line. The measured periodicities of superlattices vary from 131.5 Å for SL 1 to 134.0 for SL 3 and are consistent with the intended period of 127.8 Å (Table 1), indicating a good control over the growth rate. Differences
between the designed and measured superlattice period were attributed to be due to the small fluctuations in the GaSb growth rates and deposition of additional 1 (SL 1) – 2 (SL 3) ML of GaSb. We propose that the fluctuations in the GaSb growth rate can be due to the changes in the Sb flux and the use of a higher Sb flux resulted in a small increase (∼3%) of GaSb growth rate.

Cation and anion sublattices of SL 1 and SL 3 were imaged using cross-sectional STM as shown in Figure 3. Isovalent impurities were recognized in both anion and cation images due to both electronic and geometric effects [18]. For example, Sb for As substitution in InAs ($Sb_{As}$) appears bright relative to the surrounding As atoms because of the replacement of an As dangling bond with Sb one together with the replacement of a shorter “InAs-like” back bond with a longer “InSb-like”. In for Ga substitution in GaSb ($In_{Ga}$) likewise appears bright due to a combination of dangling-bond orbital energy and back-bond geometry [18-20].
Figure 3. Individual anion and cation sublattice images of InAs/GaSb superlattice in (110) cross-section. Sb\textsubscript{As} and In\textsubscript{Ga} denote substitutional impurities associated with segregation at the normal and inverted interfaces, respectively. Growth direction is toward the top of the page.

Compositional profiles created by In segregation at the inverted interface and by Sb segregation at the normal interface were obtained by analyzing independent, large-area surveys of anions and cations that yielded statistically significant sampling. The spatially inhomogeneous composition in the growth direction was parameterized by a segregation coefficient, R, that systematically partitioned an initial seed fraction, \( x_i \), leaving behind a monotonically decreasing impurity fraction, \( x(n) \), in successively buried layers [20].

Table 1. Parameters of the InAs/GaSb superlattices

<table>
<thead>
<tr>
<th>Superlattice</th>
<th>Incident/Consumed</th>
<th>( \Delta \alpha / \alpha_0 ) (%)</th>
<th>( \alpha_L ) (Å)</th>
<th>( L ) (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Designed</td>
<td></td>
<td></td>
<td></td>
<td>127.8</td>
</tr>
<tr>
<td>SL 1</td>
<td>1.6</td>
<td>-0.20</td>
<td>6.084</td>
<td>131.5</td>
</tr>
<tr>
<td>SL 2</td>
<td>4.5</td>
<td>-0.16</td>
<td>6.086</td>
<td>132.8</td>
</tr>
<tr>
<td>SL 3</td>
<td>8.5</td>
<td>-0.11</td>
<td>6.089</td>
<td>134.0</td>
</tr>
</tbody>
</table>
Therefore, the In segregation profile is governed by the equation:

\[ x(n) = x_i \ast (1 - R) * R^{n-1}. \]  

(A.2)

The results of the analysis are shown in Figure 4 and summarized in Table 2. The In seed is around 1 ML and \( \frac{2}{3} \) ML of In is segregating from the interface into the bulk GaSb layer. This process disturbs InSb-like termination, that was obtained through Sb-soak, and leaves a mixed predominantly GaAs-like interface behind (\( \frac{2}{3} \) GaAs-like and \( \frac{1}{3} \) InSb-like).

The presence of a residual Sb vapor background during growth of InAs introduces a non-vanishing impurity fraction, \( x_0 \), due to anion cross incorporation [19]. Therefore, the Sb impurity profile is governed by the equation:

\[ x(n) = x_i \ast (1 - R) * R^{n-1} + x_0 \ast (1 - R^n). \]  

(A.3)

The results of the analysis are shown in Figure 5 and summarized in Table 3. It is evident that Sb seeds are identical and independent of the Sb flux employed during growth. Sb seeds, that are around \( \frac{1}{3} \) of ML, are also considerably smaller.

<table>
<thead>
<tr>
<th>SL 3</th>
<th>SL 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>( x_i )</td>
<td>0.96 ± 0.02</td>
</tr>
<tr>
<td>( R_1 )</td>
<td>0.67 ± 0.01</td>
</tr>
<tr>
<td>( R_{n&gt;1} )</td>
<td>0.61 ± 0.001</td>
</tr>
</tbody>
</table>

Table 2. Model fit parameters for In segregation into GaSb layers

<table>
<thead>
<tr>
<th>SL 3</th>
<th>SL 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>( x_i )</td>
<td>0.33 ± 0.02</td>
</tr>
<tr>
<td>( R )</td>
<td>0.65 ± 0.02</td>
</tr>
<tr>
<td>( x_0 )</td>
<td>0.010 ± 0.001</td>
</tr>
</tbody>
</table>

Table 3. Model fit parameters for In segregation into GaSb layers
than In seeds, demonstrating that Sb segregation plays less significant role in advancing unintended ternary alloy formation in these structures. These seeds are also consistent with prevailing knowledge concerning the Sb surface reconstructions. Under typical Sb-rich conditions, the growth surface of GaSb is terminated with Sb bilayers, whose maximum stoichiometric excess is \(2/3\) of a ML [21]. If half of the top-layer Sb atoms are displaced by As (replacing every Sb–Sb surface dimer with a mixed As–Sb one), only a \(1/3\) ML segregation seed will remain.

From Table 1 it is evident that with the increase of the Sb flux the lattice mismatch between the InAs/GaSb superlattice and GaSb substrate is decreasing. Since the periodicity of superlattice remains nearly constant and the segregation profiles do not depend on the Sb flux, this shift in the 0th order peak position can be only explained by the change in the interface bonds. It is reasonable to suppose that the As-for-Sb exchange at the normal interface is sensitive to Sb flux. Then, a fixed segregation seed can be due to the fact that top-layer As-for-Sb replacement is quenched once the surface is terminated with mixed As-Sb dimers and that further exchange involves the supporting InSb monolayer. This implies a corresponding loss of InSb-like character at the heterojunction together with a decrease in the lattice constant of the epilayers.

The experimental X-ray diffraction pattern was analyzed with a dynamic X-ray simulation program to probe this assumption. Structural parameters used for the X-ray simulation are shown in Table 4. Sb and In impurity profiles, obtained using cross-sectional STM of SL 1 and SL 3 structures, were used to estimate the amount of In segregated from the inverted interfaces into the GaSb layers and the amount of Sb segregated from the normal interfaces and absorbed from the background into InAs layers. No STM characterization of SL 2 was performed but, based on the fact that segregation profiles were independent of Sb flux, similar values were used. The adjusted parameters were the magnitude of As-for-Sb exchange at the normal interfaces.

Figure 6 shows the comparison between the experimental and calculated (400) diffraction patterns for three structures studied. The best fits were obtained with As-for-Sb exchange set at 0.27% for SL 1 (resulting in the mixed interface that is \(3/4\) InSb-like and \(1/4\) GaAs-like), 0.13% for SL 2, and 0% for SL 3 (resulting in the perfect InSb-like interface). The positions and maximum reflectivity of

<table>
<thead>
<tr>
<th>Segregated In (monolayer)</th>
<th>Background Sb (monolayer)</th>
<th>Segregated Sb (monolayer)</th>
<th>As-for-Sb exchange (%)</th>
<th>L (Å)</th>
<th>Simulation</th>
</tr>
</thead>
<tbody>
<tr>
<td>STM</td>
<td>Simulation</td>
<td>STM</td>
<td>Simulation</td>
<td>STM</td>
<td>Simulation</td>
</tr>
<tr>
<td>SL 1</td>
<td>0.64</td>
<td>0.64</td>
<td>0.006</td>
<td>7.0000e-3</td>
<td>0.33</td>
</tr>
<tr>
<td>SL 2</td>
<td>0.64</td>
<td>0.006</td>
<td>8.0000e-3</td>
<td>0.36</td>
<td>0.1300</td>
</tr>
<tr>
<td>SL 3</td>
<td>0.64</td>
<td>0.010</td>
<td>0.010</td>
<td>0.31</td>
<td>0.36</td>
</tr>
</tbody>
</table>
the 0th order peak and satellite peaks are in excellent agreement with the measured patterns, supporting our assumptions. Therefore, the evolution in the zero 0th order peak position was successfully tracked by increasing the GaAs-like character at the normal interface while keeping the InAs layer thickness, and inverted heterojunction composition fixed. These results also indicate that high Sb flux, used during the GaSb growth, helped to prevent In-Sb type bonding at the normal interface while unintended As-for-Sb exchange occurred when a low Sb flux was used. This suggests that particular care must be exercised over unintended As-for-Sb exchange in order to properly control bond-type selectivity, as well as strain balance within these heterostructures.

4 Conclusion

In summary, we demonstrated that DCXRD coupled with cross-sectional STM can be used for accurate and reliable structural analysis of InAs / GaSb superlattices. They provide detailed information on the thickness and composition of the alternating layers as well as on the interfaces between them. It was observed that cation segregation at the inverted interface disturbed InSb-like termination resulting in a mixed predominantly GaAs-like interface. The effect of Sb flux
on the structural properties of the superlattice was studied. Although In and Sb segregation profiles did not depend on the Sb flux, high Sb flux helped to minimize As-for-Sb exchange at the InAs-on-GaSb interface and to preserve In-Sb type bonds.

References