Self-Organized Superlattices in GaInAsSb Grown on Vicinal Substrates

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ABSTRACT

Self-organized superlattices are observed in GaInAsSb epilayers grown lattice matched to vicinal GaSb substrates. The natural superlattice (NSL) is oriented at a slight angle of about 4° with respect to the vicinal (001) GaSb substrate. This vertical composition modulation is detected at the onset of growth. Layers in the NSL are continuous over the lateral extent of the substrate. Furthermore, the NSL persists throughout several microns of deposition. The NSLs have a period ranging from 10 to 30 nm, which is dependent on deposition temperature and GaInAsSb alloy composition. While the principal driving force for this type of phase separation is chemical, the mechanism for the self-organized microstructure is related to local strains associated with surface undulations. By using a substrate with surface undulations, the tilted NSL can be induced in layers with alloy compositions that normally do not exhibit this self-organized microstructure under typical growth conditions. These results underscore the complex interactions between compositional and morphological perturbations.

INTRODUCTION

Phase separation in multi-component compound semiconductors has been widely reported [1]. Of particular interest are materials systems that spontaneously self-organize with a significant degree of regularity, since this periodicity can impact the electronic band structure and consequently, materials properties and device performance. The length scale of these ordered phases ranges from the atomic scale, e.g. CuPt ordering as observed in GaInP [2], GaAsP [3], GaAsSb, [4], and InAsSb [5], to microscopic dimensions on the order of ~50 nm, e.g. composition modulation. Composition modulation can persist either parallel (lateral) to the growth direction, or perpendicular (vertical) to the growth direction. Lateral composition modulation (LCM) has been reported in strained alloy systems such as bulk AlInAs and GaInP epilayers [6-8], as well as in short period superlattices such as GaP/InP, AlAs/InAs, GaAs/InAs, and InAs/GaSb [9-12].

Vertical composition modulation (VCM) and self-organized natural superlattices (NSLs) in alloy layers that are homogeneously grown have also been observed, but these studies are less prevalent and the mechanism for their formation are less understood. NSLs have been reported in ZnSeTe grown on vicinal GaAs substrates [13] and SiGe grown on (001) Si [14]. The NSL period was 2 to 3 nm for both of these materials systems, and a model based on step-flow growth and local strain fields that are modulated during growth were developed to explain the phenomena [14]. In addition, InAsSb and GaAsSb [5,15] were reported to spontaneously form a periodic structure that consisted of platelets of alternating composition and periodicity on the order of 20 to 50 nm. Both InAsSb and GaAsSb exhibit miscibility gaps [16], and this larger scale modulation was attributed to the tendency for these alloys to phase separate. It was speculated that islands of the different phases develop at the growth surface and then

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and was used to correlate the microstructure with the surface undulations. The nanophotolithography was characterized by atomic force microscopy (AFM) operating in tapping mode. Quantitatively determining the area of feature lines, the period of the line (TNS), the surface
in addition, HRXRD rocking curves and reciprocal space X-ray maps (RSeXRD) were used to
imaged using £ = 222 or £ = 131. The beam conditions, in which bright-field dark-field conditions
Scanning electron microscopy (SEM) and TEM operated at 200 kW. For TEM, the NCSL was
GaN/AlGAs was studied by examining 1-10 and 1-11 cross-sections in at-field-ermission
Several methods were used to characterize the CAs and NCSL. The microscope of
were used to assess alloy non-uniformity [2].

EXPERIMENTAL APPROACH

increase

to the insubility gap, both intensity of the NLS and amplitude of surface undulations

growth period is correlated with the wavelength of surface undulations on the

RELS, 20 also reported that no NLS were observed in either GaN/AlGaAs or AlGaAs.
the NLS base modulation period ranging from 10 to 20 nm. It is interesting to note, however, that

The common trend in these reports on VCM is that each of the alloys studied exhibits a

GaN/AlGaAs. In this study, the alloys were also grown on a silicon substrate and dark and light
crapping. [17] In this study, the alloy was also grown on a silicon substrate and dark and light

subsequently laterally overgrow each other. While atomic ordering and VCM were
These results indicate that the initial microstructural mismatch of 4° will affect the angle of the CSLs and additional samples grown under various conditions can be used to characterize the effect of CSLs and additional samples grown under various conditions.

The first CSL can also be characterized by RAXM. Figure 4 and 5 show a HAXRD.

The geometry of the microstructure is shown in Figure 4 and 5. The micrograph shows a cross-section of the TEM, which was taken from the TEM imaging of the sample. The HAXRD image shows the contrast between the CSLs and the additional samples grown under various conditions.

The results show that a variety of techniques, including TEM, FE-SEM, and HAXRD can be used to characterize the initial CSLs and additional samples grown under various conditions.
The effect on alloy composition on the intensity of the tilted microstructure was determined by RIXRM, and Figure 6 shows these maps for three different alloy compositions grown on (001) GaSb miscut 6° toward [1-10]. The 300 K PL peak wavelength of the samples are 2.09, 2.07, and 2.485 μm, for Figs. 6a, 6b, and 6c, respectively. The intensity associated with the tilted microstructure increases with increasing PL peak wavelength, i.e., as the alloy composition moves further into the miscibility gap.

Figure 7a is GaInAsSb grown at 525 °C on (001) GaSb miscut 6° toward [1-11]B and Fig. 7b is GaInAsSb grown at 575 °C on (001) GaSb miscut 2° toward [011]. A surface undulation is observed for both of these images, and it is characteristic of all the samples that exhibited the tilted microstructure. Figure 7c shows the step-bunched 575 °C [23] and the sample shown in Fig. 7b exhibits both the step bunching and surface undulations.

The wavelength of these surface undulations is directly correlated with the period of the tilted NSL. The surface undulation of the AFM image shown in Fig. 7a has a period of about 11.5 nm. The tilted NSL has a period of 20 nm and a tilt angle of 10°. Simple geometry indicates that when a 20 nm period tilted at 10° intersects the surface, it will have a lateral period of 20 nm, as 115 nm. Thus, the lateral period of the tilted NSL is larger for a smaller misorientation angle. Furthermore, it was found that the amplitude of the undulation increases with the strength of the tilted NSL, as observed in RIXRM. The amplitude of the undulation is less than 1 nm for the sample shown in Fig. 6a, while it is 4-5 nm for the sample shown in Fig. 6c.

These results suggest that the tilted NSL and surface undulations are coupled. To further test this hypothesis, a structure was specifically grown. The layer structure is shown in Figure 8. It consists of different alloy compositions of GaInAsSb, separated by 2 nm GaSb. The alloy composition of layer #1 and #5 is similar to that of the layer shown in Figs. 1b and 2. Therefore, the observations of a tilted NSL and strong spinodal-like contrast are anticipated in layer #3, but not in layers #1 and #5.

As expected, the <220> image (Fig. 9a) indicates that layers #1 and #5 barely exhibit more contrast than the GaSb substrate, while layer #3 exhibits strong spinodal-like contrast. Figure 9b shows a <222> beam image of the substrate, buffer layer, and layers #1-3, while layer #5, no NSL is observed. No NSL is observed in layer #5 (Fig. 9c). However, a tilted NSL is observed throughout the entire epitaxial layer, although it appears to weaken as growth proceeds.

The tilted NSL present in layer #3 was propagated into layer #5, even though the NSL was not present in layer #1 and layers #1 and #5 were grown under the same temperature and flow conditions. These images clearly illustrate the coupling of compositional and morphological perturbations [26-29]. The composition associated with layer #5 does not inherently phase...
a similar mechanism to that in the Vesicular model. Rather than occlusion and capture of single

The additional life of the supernatant with respect to the surface steps can be understood from

surface reconstructions (chimeras) [13].

seen in many III-V semiconductors, which is now believed to be caused by
multiple layers of quantum dots [30], whereas another is the growth of a single.

The strain-locking mechanisms due to surface strain are known
due to the strain field of the surface undulations. Such strain-locking explains the
inspirations of the suppression. Such strain-locking explains the

Once such undulations form, all subsurface defects disappear when each successive

AVFM images in Fig. 7.

The surface forms a sea of peaks and valleys, creating surface undulations. As are observed in the

A qualitative model for development of the VBM is proposed here. The model is based on the

proposed that surface undulations play a similar role to step bunches in the Vesicular model.

suggests that surface undulations are directly related to the VBM. This

because the surface undulations are directly related to the VBM. The VBM is directly correlated to the

These observations are in line with models developed by Vesarela et al. [14], which

DISCUSSION

surface reconstructions, and shows that surface strain/roughness can promote phase separation.

Therefore, the VBM cannot explain the larger NSL periods observed

not GaAs, but GaAs, which have the surface undulations. For example, the undulations

The lateral extent of the VBM is about 90 nm, and a height of 7.7 Å. Voids in a lateral period of

separate to form a liquid NSL. It did so in this special case because surface undulations present
REFERENCES

ACKNOWLEDGMENTS

Conclusions

composition of the liquid NSL is discussed. A qualitative model for the propagation and modulation of multifunctional perturbations is considered. The authors gratefully acknowledge PM Nilshin and J.W. Chudzik for technical assistance in materials characterization.

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<table>
<thead>
<tr>
<th>Width (mm)</th>
<th>NSL Period</th>
<th>NSL Til Angle</th>
<th>HFM Period</th>
<th>Miscellaneous</th>
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<tr>
<td>160</td>
<td>132</td>
<td>6.0°</td>
<td>13.8°</td>
<td>1.0° [101]</td>
</tr>
<tr>
<td>143</td>
<td>144</td>
<td>5.8°</td>
<td>14.6°</td>
<td>1.1° [111]</td>
</tr>
<tr>
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<td>115</td>
<td>1.0°</td>
<td>20.0°</td>
<td>2.2° [111]</td>
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Table 1. Correlation of NSL Lateral Surface and Surface Undulations
Surface normal, i.e., 4° in addition to the 6° miscut angle.

Conditions: Spindel-like contrast is observed as well as a NSL. The NSL is tilted 10° from the 222<2-Beam.

Figure 2. [110] Cross-section TEM image of GaAs/InAs$_{0.25}$Sb$_{0.75}$ sample using $\theta = 222<2$-Beam.

Figure 3. Bright-field [1-10] cross-section TEM images with $\theta = 220<2$-Beam direction of GaAs$_{0.25}$Sb$_{0.75}$ grown at 725°C on substrates oriented (011) 6° off (1-11): (a) GaAs$_{0.25}$Sb$_{0.75}$ and (b) GaAs$_{0.25}$Sb$_{0.75}$ with GaSb.
Figure 4. A schematic diagram of the self-organized NSL microstructure.

Figure 3. Cross-section FE-SEM images of CaSb$_2$O$_3$ showing the NSL: (a) [1-1-10] cross section and (q) [1-1-10] cross section.
Figure 6. Rietveld refinement of (a) \(Ca_{0.8}In_{0.2}Sb_2\) and (b) \(Ca_{0.8}Sb_{1.2}\) with the Rietveld method.

Figure 5. HR-XRD of \(Ca_{0.8}In_{0.2}Sb_2\): (a) 0/2θ scan and (b) reciprocal space map.
Figure 8. Schematic structure of specifically grown GaN/AlN film. To observe coupling of morphological surface undulations and compositional modulation.

Figure 7. AFM images of the surface undulations of GaN/AlN samples with a tilted [11-1] (100) (a) and [10I] (100) (b) superlattice grown on various GaN substrate misfit orientation: (a) (100) 6° rotated B and

\[ \text{[11-1] (100)} \]