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Relaxation and tilting of single and double layer structures of AlGaSb/GaSb-LPE studied by high resolution x-ray diffraction

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Abstract

Epitaxial AlGaSb double-layer structures grown on GaSb (001) substrates by Liquid Phase Epitaxy (LPE) were analyzed by high-resolution x-ray diffraction (HRXRD). Four AlGaSb double-layer structures grown at 450 °C were analyzed varying the thickness of the first layer and maintaining the same thickness for a second layer growth. Symmetric reciprocal space mapping measurements around the (004) reflection and asymmetric rocking curves around the (115) reflections have revealed that a subsequent Al0.15Ga0.85Sb growth on an Al0.047Ga0.953Sb layer modifies the relaxation and lattice tilting of the first layer. This behavior is attributed to the formation of dislocations within the layers during the growth and transported between them. In this work, the study was realized ex situ and is in well agreement with in situ studies and theoretical predictions on the relaxation of epitaxial films in diverse materials. The structural analysis of lattice distortion in epitaxial layers is relevant since it could modify the electrical behavior of optoelectronic devices building with them.

1. Introduction

The AlxGa1−xSb alloys are materials with potential applications in optoelectronic devices operating in the wavelength range from 1.3 to 5 μm, such as avalanche photodiodes [1], light emitter diodes [2] and lasers [3, 4]. This alloy can be adjusted between 0.72 and 0.93 eV as a direct bandgap semiconductor by varying its composition from x = 0 to 0.4, in this range the alloy can tuned to operate at the wavelength with less attenuation in optical fiber at 1.55 μm [5]. However, in order to improve the performance of the optoelectronic devices, the development of high quality layers grown on substrates is mandatory [6, 7].

Despite the relative small lattice mismatch between AlxGa1−xSb layers and GaSb substrates (4.9 × 10−4 for x = 0.047 and 1.6 × 10−3 for x = 0.15) and difference of thermal expansion coefficient (0.087 × 10−6 K−1 for x = 0.047 and 0.37 × 10−6 K−1 for x = 0.15) [8, 9], the heterojunction presents strain when the thickness exceeds the critical value. Under controlled conditions is possible to grow an epitaxial AlGaSb layer matched to a GaSb substrate until the thickness of layer reaches the critical value, [10], which depends on the aluminum content, x, and the growth temperature, Tg. If the critical thickness is exceeded, the layer it becomes energetically favourable for the strain to be relieved through the formation of dislocations at the interface. In order to design layered structures with controlled strain, it is necessary to know the structural state in each constituent layer, and whether this is affected by the subsequent layers in the structure. Also, the tilt in epitaxial layers has been shown that modifies the electrical parameters of some devices grown on tilted substrates [11].

Reciprocal space mapping (RSM) is a useful tool to investigate the structural characteristics of epitaxial thin films and thus achieve a deeper understanding of the strain domain [12], which shows the change from the fully strained to the relaxed state into the layers grown on single-crystal substrates [13]. However, to our knowledge,
this technique has not been used to analyze the effect of subsequent growth of AlGaSb layer on GaSb substrates within the thickness range presented.

In this work, we analyze through symmetric RSM and asymmetric rocking curves, the relaxation and tilting of single and double layer structures of AlGaSb/GaSb grown by liquid phase epitaxy (LPE) on GaSb (100) substrates. The double layer structure is intended to be used as the basis of a photodetector for optical fiber communications at 1.55 μm. The first layer grown on the substrate is used as the active material for the light absorption and the second layer serves as a window for transmission of light, the thickness of the first layer was increased beyond the critical value to enhance the photon absorption.

A set of double layer structures with the same Al composition and different first layer thicknesses were compared to a similar set of single layer structures. The comparison is done in order to determine the effect on the relaxation and tilt in the first layer due to the growth of a second layer on it. This type of study has been performed before in situ during growth stage [14] and exist theoretical calculations about the plastic relaxation of multilayer structures [15]. The study was realized ex situ, however, is according with in situ studies and predictions of relaxation in related materials.

2. Experimental methods

The double structures were made by two successive growths of AlxGa1-xSb (x = 0.047 and 0.15) layers on GaSb (100) substrates using a typical Liquid Phase Epitaxy (LPE) system inside a horizontal quartz furnace under a hydrogen environment at 450 °C. The layer growth was originated from two melts composed of 6 N Ga-rich mixed with 6 N Al and 5 N GaSb. In order to obtain a concentration of Al x = 0.047 and x = 0.15 for the first and second layer, respectively, the composition of the melt was determined using the thermodynamic parameters and calculations for AlGaSb, published by Elyukhin [16].

The procedure used to grow the layers inside the furnace was as follows: the melt solutions were homogenized at 600 °C for two hours, then, the temperature was lowered to 450 °C in order to reach the equilibrium condition for the first layer during one hour, after that, the melts were supercooled lowering the temperature with a cooling rate of 0.5 °C min⁻¹ until 10 °C below the equilibrium temperature, thus, the first solution and substrate was brought in contact during the different growth times: 1, 2, 3 and 4 min. The second layer growth was initiated after separates the first solution from substrate and immediately brought in contact with the second solution, the composition of the second melt were calculated accordingly to temperature reached at this time, keeping the supercooling for the first melt, the growth time for the second layer was 2 min for all structures.

Thicknesses of layers were measured using a scanning electron microscope (SEM) JEOL JSM-6610LV, by identifying the interfaces in lateral images. High resolution x-ray diffraction rocking curves and RSMs from multilayers were recorded employing a Bruker D8 Discover diffractometer using the CuKα1 radiation (λ = 1.54056 Å) obtained from a V-groove Ge compressor as a monochromator. The rocking curves obtained were symmetric and asymmetric for the (004) and (115) reflections, respectively. Reciprocal space maps (RSMs) around the GaSb (004) reflection were acquired using a point detector to obtain a suitable resolution.

The double structures were used to fabricate photodetectors tuned at 1.55 μm, the responsivity was measured in order to determine the more suitable first layer thickness to improve the device performance.

3. Theory

Relevant information about the structure of epitaxial layers grown on a substrate is contained in the rocking curves and the RSM obtained by high resolution x-ray diffraction. This information is extracted primarily from peaks position of layers relative to the substrate, one of the structural issue obtained by this analysis is the relaxation degree in layers from fully strained to fully relaxed. The degree of relaxation \( R \) is defined from the lattice parameters of the substrate and the layer:

\[
R = \frac{a(L) - a_0(S)}{a_0(L) - a_0(S)}
\]

where \( a(L) \) and \( a_0(L) \) are the measured and relaxed lattice parameters, respectively, of a layer of given composition, and \( a_0(S) \) is the relaxed lattice parameter of the substrate.

Another structural issue that could be observed in the peak positions of the RSM is the misorientation or tilt of layers respect to the substrate as already documented by other researchers [17, 18]. In order to calculate the tilt \( \gamma \) in layers were used their reciprocal space coordinates in the Qx and Qz direction, according to the formulas published by Roesener [19] and geometrical calculations of Chauveau [20]:
\[ \tan(\gamma) = \frac{\Delta Q_x^{(004)}}{2\lambda/a_s - |\Delta Q_z^{(004)}|} \]  

(2)

where \( \Delta Q_x^{(004)} \) and \( \Delta Q_z^{(004)} \) are the coordinates of layer respect to substrate in the RSM (004) in x and z direction, respectively, \( \lambda \) is the wavelength used in measurement, and \( a_s \) is the substrate lattice parameter. 

Besides the miscut of the substrate, another cause of tilt in layers is the generation of dislocations in the interface due to the relaxation process in the layer [21]. Estimation of dislocation density \( N_S \) by x-ray diffraction can be expressed as follows [22]:

\[ N_S = \frac{a_{\omega}^2}{4.36b^2} \]  

(3)

where \( a_{\omega} \) is the FWHM of the \( \omega \) scan and \( b \) is the burgers vector. Figure 1(a) shows a sketch of the double-layer depicting a tilt \( \gamma \) respect to the substrate, figure 1(b) outlines the layer peak displacements in the RSM due to change of relaxation and/or tilt. The parallel and perpendicular parameters were calculated from asymmetric rocking curves from 115 reflections using the formulas developed by Macrander [23].

4. Results

The measurement of layers thicknesses was performed using SEM micrographs due to dimension order of layers, a transversal section image of the A2 sample is showed in figure 2, where can be observed the interfaces between the substrate and the first and second layer.

Asymmetrical rocking curves for (1-15) and (1 15) reflections of the double layer structures are presented in figure 3, here can be identified the peaks for the substrate and for the two layers, also, can be noticed the change in the positions of layers peaks as the first layer thickness is increased. The peaks shift is attributed to lattice constant change originated by relaxation process in the layers and the intensity decreasing is originated by the misorientation of crystalline structure. 

Projections in reciprocal units of the (004) RSMs from double-layer structures for the different growth times and thickness of the first layer are shown in figure 4. In the RSMs can be observed three maximums corresponding to the peaks of substrate, first layer, and second layer. The peak position of the layers moves along the Qz direction as the thickness is increased, towards the substrate peak, this is attributed to the structure relaxation as the first layer thickness is increased. The peaks of both layers also shift along the Qx direction as the first layer thickness is increased, this corresponds to an increment in lattice tilting for both layers respect to substrate. Also, it can be observed scattering around the three reflections which is changed as the thickness is increased, this behavior corresponds to dislocations generated at the heterointerface accordingly to reported by other authors [14, 24].
In a previous work [25], structural characterization of single-layer AlGaSb structures grown on GaSb substrates with growth conditions similar to the first layer of the double structure was reported. The values of relaxation and tilting of the single-layer are compared with the double-layer structure in order to reveal the effect of the second growth and show an interaction between the layers.

The relaxation values for single and double layer structures were calculated from asymmetric (115) rocking curves, these are presented as function of the first layer thickness in figure 5, here, the critical thickness $h_c$.
calculated using theory published by Matthews and Blakeslee [10] is placed as reference. Regards to single layer structure when the thickness is below the critical value (≈2.9 μm) a layer virtually without relaxation was grown, as the thickness is increased the relaxation increases rapidly from a fully strained state to fully relaxed state. However, in two-layer structures the first layer relaxation presents noticeable increment compared to single-layer structure, despite the thickness values are similar for both structures. The highest increment is for thickness below the critical value, around 70%, this increment is reduced as the thickness is increased, which indicates that the second layer produces relaxation in function of the strain present in the layer.

Figure 6 shows the lattice tilt respect to the substrate calculated from Qx displacement in RSMs for the layers in the single and two-layer structures. In the figure can be observed a rising of tilting for the single layer as the thickness is increased, regards to the first layer in the double structure a similar behavior is observed but with an increment nearly constant respect to single layer, also, is not observed a trend to reach a maximum value.
The calculated tilt for the second layer shows an approximately constant shift with respect to the first layer tilt, presenting a very similar behavior in function of thickness, the analogous increment observed in the first layer compared to single-layer revealed the effect of increasing the tilt due to the subsequent second layer grown on this, and that the increment of tilt is result of interaction between the layers.

Estimation of dislocation density is showed in the table 1, where can be observed an increase of density in the first layer when the second layer is grown on, also the estimation of dislocation density in the second layer is near one order of magnitude higher than dislocations in the first layer.

Responsivity measurements at 1.55 μm obtained from photodetectors fabricated with the double structures are presented in figure 7, here can be observed that the best performance is from the device fabricated with the structure that presented a moderated relaxation and tilt.

5. Discussion

The interaction between the layers of a double AlGaSb layer structure grown by LPE has been showed by the presented results. The first effect is observed the position shift of peaks in asymmetrical rocking curves which indicates a significant increase of the first layer relaxation by the growth of the second layer compared to value in a single layer structure.

Another effect of the interaction is observed in the maximum shift of layers in the reciprocal space maps which indicates a tilt in the first layer after the second layer is grown, this increment is near the same for all samples with different first layer thickness, attributed to the unvarying growth time for the second layer.

A consistent explanation for mechanism that increases the relaxation and tilt of the first layer is that the second layer growth is producing an increment of strain in the entire structure, which in turn creates dislocations, spreading them from the interface between layers towards the substrate interface, when dislocations reach this interface generate more dislocations [26, 27]. Estimation of dislocation density is agreed with this, because the values are increased in both layers.

The behavior of relaxation and tilt values with the thickness also reveal the interaction between the layers, the most remarkable features are described below. The increment in the relaxation rate for the first layer in the
double layer structure compared to single layer is attributed to the strain release caused by dislocations generation when the second layer is grown, for thickness above 4 $\mu m$ a decrease in the relaxation rate of the second layer meanwhile an increase in the relaxation rate in the first layer is observed, this is attributed to the second layer is grown on a material with larger lattice parameter and the mismatch is lower.

The tilting increment of the first layer in the double structure is nearly a constant value for all the samples compared to the single structure, which indicates that the generation of dislocations that causes the tilting in the first layer are under the same conditions during the second layer growth. The tilting value does not show a trend to reach a maximum value as is observed for relaxation, which indicates that the motion of dislocations continues even after reaching the full relaxation.

The comparison of the performance of the devices with different first layer thickness was obtained measuring the responsivity of the structures, observing that a moderate relaxation with a thickness close to the critical improves their response, however, increasing the thickness increases photon absorption but the consequent increase in relaxation and tilt of the layers decreases the spectral response of the device.

6. Summary

The relaxation and tilting behavior in an AlGaSb double-layer structure compared to single layer structure grown on a GaSb substrate by the LPE technique is presented. The study was realized using asymmetric rocking curves and symmetric reciprocal space maps. The results revealed a notable increment of the relaxation and tilting in the first AlGaSb layer due to the growth of a second AlGaSb layer on it, and a consistent cause is the generation and motion of dislocations between the layers. A comparison of device performance fabricated with the double structures was realized by responsivity measurement at 1.55 $\mu m$.

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