Crystal lattice tilt analysis in gradient composition layers by electron and X-ray diffraction

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Abstract. The combination of X-ray tilt-azimuth figure, reciprocal space map and transmission electron microscopy in selected area electron diffraction mode was applied to analyse the tilt of the MBE-grown 1.2-µm thick convex-graded InxAl1-xAs (x=0.05-0.80) metamorphic buffer layer on singular GaAs (001) substrate. The combination of the techniques is demonstrated to enable the determination of the tilt angle as well as the tilt direction together with monitoring of the tilt development along the growth direction. For the studied metamorphic buffer layer as an example, the tilt direction was deduced to be 20° off the (110) GaAs crystallographic plane. The tilt angle of (002) buffer layer planes was observed to rapidly rise to 0.6° near the layer-substrate interface and then to slowly increase to 1.4° with the approach toward the surface.

1. Introduction

The epitaxial growth of perfect semiconductor solid solutions is possible only within the narrow composition range due to restricted set of lattice constants provided by available elemental or binary substrates. To grow device structures of which the lattice constant does not match that of the available substrates, metamorphic buffer layers (MBL) are used as a transition region between the substrate and an active structure [1-4]. Due to an essential difference in the lattice constant between the substrate and MBL, arising mismatch strain necessarily relaxes via generation of misfit dislocations. The MBL design and growth conditions have to be optimized as to achieve the complete mismatch strain relaxation without propagating the dislocations into the upper-lying active region.

The key issue in the epitaxial deposition of MBL is the control of the mismatch strain and its relaxation which detrimentally affects the electronic properties. For the general characterization of the strain relaxation of MBL, X-ray reciprocal space mapping (RSM) is widely used [5, 6]. The diffracted intensity distribution around a reflection plotted on the RSM is sensitive to the composition of MBL, to the accumulated strain and to the MBL tilt. Unfortunately, for the compositionally graded MBL with nonuniform strain relaxation along the growth direction (i.e. over the layer thickness), correlation of the RSM intensity distribution to the depth scale becomes questionable. Space resolved information on the strain state, defect types and distribution along the layer thickness can be provided by transmission electron microscopy (TEM) [7-9].

For the analysis of the strain relaxation in compositionally graded mismatched layers, combined RSM and TEM data representation in the form of RSM with superimposed lattice parameter profiles (m-profiles) measured by cross-section TEM was shown to be very informative [10]. The profiling procedure was applied to about 1-µm thick linearly-graded InxGa1-xAs (x=0.05-0.38) [11] as well as to
1-µm thick convex-graded In\textsubscript{x}Al\textsubscript{1-x}As (x=0.05-0.79) [12] MBLs, both MBE-grown on the GaAs (001) substrate. The RSM with plotted m-profile was designated as the depth profiled RSM (DpRSM).

In conjunction with the mismatch strain relief through the dislocation generation and propagation, a tilt in mismatched layers occurs if dislocation densities differ in glide systems [13]. In this paper we apply DpRSM and additionally involve tilt-azimuth figure (TAF) recording to monitor the tilt behavior in 1.2-µm thick convex-graded In\textsubscript{x}Al\textsubscript{1-x}As (x=0.05-0.80) MBL.

2. Experimental

The sample containing convex-graded In\textsubscript{x}Al\textsubscript{1-x}As MBL was grown by molecular beam epitaxy on semi-insulating GaAs (001) substrate. The growth procedure is described in more detail elsewhere [12]. The complete layer sequence of the sample under study is presented in Fig. 1.

![Figure 1](image)

**Figure 1.** Design of the sample under study with In\textsubscript{x}Al\textsubscript{1-x}As (x=0.05-0.8) metamorphic buffer layer on GaAs (001) substrate.

High-resolution XRD measurements were carried out using multifunctional triple-crystal diffractometer D8 Discover (Bruker, Germany) in a parallel X-ray beam geometry. The $\theta$ - 2$\theta$ scan as well as RSM around symmetric (004) GaAs reflection were used for the analysis of crystalline properties of the MBL. The TAF was recorded by collecting intensities during $\omega$-scans at different azimuthal angles $\varphi$.

Cross-section specimens for TEM were prepared by conventional procedure of mechanical grinding-polishing and final Ar$^+$ ion milling.

For TEM study, JEM2100F electron microscope operating under an accelerating voltage of 200 kV was exploited in the conventional modes of diffraction contrast imaging and selected area electron diffraction (SAED).

3. Results and Discussion

The TAF from the InAlAs MBL was collected at the angle 2$\theta$ = 64.6°, which corresponds to the diffraction conditions for In\textsubscript{0.7}Ga\textsubscript{0.3}As composition. The correlation of the TAF collection region with the layer depth scale can be seen in Fig. 2. The geometry of the TAF recording is shown in Fig. 3a. During the experiment the sample was positioned in such a way that [1\overline{1}0] axis of GaAs substrate coincided with the azimuthal direction to 0° of the figure.

The collected TAF is represented in Fig. 3b. The diffuse intensity cloud about 0.5° wide clearly appears within the azimuthal angle range 90°-270°. At the azimuthal angles 0-90° and 270-360° the intensity attenuates, but the intensity behavior being still distinguishable. The intensity depletion results from a deviation of the (002) MBL atomic planes from the exact diffraction conditions due to
the presence of a tilt, of which the magnitude is greater than the rocking curve full width at half maximum $\Delta \omega$.

The measured tilt angle $\alpha$ is known to depend on the in-plane sample orientation azimuthal angle $\varphi$ in respect to the incident X-ray beam, obeying to the sinusoidal dependence $\alpha = \alpha_0 \cos(\varphi - \varphi_0)$, where $\alpha_0$ and $\varphi_0$ are the true tilt and the tilt azimuthal angles respectively [14]. Consequently, a maximal tilt angle of 0.9° deduced from the figure was accepted as the true tilt angle $\alpha_0$. It was observed at the azimuthal angles approximately 110° and 290°, i.e. 20° off the (220) crystallographic plane of the sample.

![Figure 2](image)

**Figure 2.** $\omega$-$2\theta$ - scan of the sample near (004) GaAs reflection. The section, which the TAF was recorded from, is marked by vertical dashed line at $2\theta = 64.6^\circ$.

To monitor the tilt behavior along the layer thickness, a set of SAED patterns registered from (110) cross-section with the selected-area aperture size as small as 100 nm was obtained shifting the sample to a regular interval of approximately 30 nm along the growth direction. Two angles describing layer lattice tilt were measured with the reference to GaAs substrate: $\alpha^\perp$ for the tilt of (002) planes and $\alpha^\parallel$ for the tilt of (220) planes. Simultaneously, the 002 and 220 diffraction vector length $q^\perp$ and $q^\parallel$ were measured, and the in-plane $m^\perp$ and out-of-plane $m^\parallel$ mismatch was derived from a simple equation $m = \Delta q / q_s = (a_l - a_s) / a_s$, where subscripts “l” and “s” stand for the MBL and substrate correspondingly. The data are represented in Fig. 4.

As it can be seen from Fig. 4, the layer tilt occurs quite close to the layer-substrate interface, its value rapidly reaches 0.6-0.8° already at a distance of 100 nm. Note that here the measured tilt angles $\alpha^\perp$ and $\alpha^\parallel$ become perceptibly different. Then, two smooth steps are noticeable in the tilt behavior. The first step appears around 300 nm from the interface (point 1 on the graph) and results in both tilt angles increasing to the same value of approximately 1.0°. The second increase in the tilt starts at
around 780 nm from the interface (point 2), and the layer tilt arrives to 1.1-1.4° at 1000 nm from the interface (point 3), both tilt angles becoming different again. It should be noticed that the azimuthal figure (Fig. 3b) is recorded from the bottom part of MBL, where In concentration reaches x=0.3, i.e. at a distance of about 300 nm from the layer-substrate interface. One more remark is that the tilt angle measured by SAED is slightly less than the true tilt angle since the tilt vector deviates 20° off the (110) plane. With this in mind, the tilt angle measured by SAED at the point 1 coincides rather well with that deduced from TAF.

Figure 3. (a) Schematic of the experimental setup geometry and (b) tilt-azimuth figure recorded at 2θ = 64.6° from a bottom part of the metamorphic buffer layer.

The combined data representation in the form of RSM and the lattice parameter profiles (m-profiles) has been shown to be a useful visual representation of the quantitative data on the distribution of the strain and composition of MBL along the growth direction [10]. We collected the symmetrical (004) DpRSM which is known to reflect purely the tilt but not the strain. The DpRSM is replicated in Fig. 5 and shows the deviation of the layer node from the surface normal (qz direction) due to presence of a tilt. The profile reflecting out-of-plane mismatch m⊥ obtained by SAED is plotted on RSM with the reference to the GaAs substrate. The m⊥ profile is represented by vertical red line in RSM and, as it can be seen, does not provide any valuable information on the mismatch behavior. The tilt correction can be introduced by comparison of the RSMs for symmetric and asymmetric reflections. However,
the procedure for collecting few RSMs and performing large data treatment is rather complex and time consuming. Instead, to account for the MBL tilt we used the data obtained by SAED. The corrected profile is represented by blue line and allows one to trace both lattice mismatch and tilt angle behavior.

**Figure 4.** Behavior of out-of-plane $m^\perp$ and in-plane $m^\parallel$ lattice mismatch and tilt angles $\alpha^\perp$ and $\alpha^\parallel$ with increasing distance from GaAs substrate.

**Figure 5.** Reciprocal space map around symmetrical 004 node of the reciprocal lattice of InAlAs metamorphic buffer layer grown on GaAs (001) substrate.
The characteristic points associated with the tilt behavior features observable in Fig. 4 are reproduced in the RSM.

4. Summary
We applied the procedure of X-ray TAF and RSM constructions combined with TEM SAED profiling to study the tilt of the MBE-grown In$_x$Al$_{1-x}$As (x=0.05-0.80) MBL in respect to GaAs (001) substrate. The combination of the techniques is demonstrated to enable the determination of the MBL tilt angle as well as the tilt direction together with monitoring of the tilt development along the growth direction. As a result, the tilt direction of 1.2-µm thick convex-graded In$_x$Al$_{1-x}$As (x=0.05-0.80) MBL was deduced to be 20° off the [110] GaAs substrate crystallographic axis. The tilt angle of (002) MBL planes was observed to rapidly rise to 0.6° near the interface and then to slowly stepwise increase to 1.4° with the approach toward the surface.

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