Structural quality and ordering of MBE grown Al$_x$Ga$_{1-x}$N-layers

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Abstract

Epitaxial Al$_x$Ga$_{1-x}$N-layers ($x = 1–0$) with the hexagonal wurzite structure were grown on 6H-SiC- and $\alpha$-Al$_2$O$_3$-substrates for the development of semiconductor UV detectors. The experiments were performed in a plasma enhanced molecular beam epitaxy system (MBE) by varying substrate temperature, III/V ratio and growth rates. A detailed analysis of dislocations was performed for pure AlN-layers by RBS-channeling and was compared to the analysis of cross sectional and plan view TEM images. The annihilation of threading dislocations during the growth process was observed by RBS-channeling depending on the type of substrate and the growth mechanism. Lowest dislocation densities were obtained for 2D-growth on SiC substrate in the range of $2 \times 10^8$ cm$^{-2}$. The decrease of the growth temperature from 1000 to 900°C lead to a decrease of the dislocation density by about an order of magnitude. Depending on the growth conditions chemical ordering was observed for ternary Al$_x$Ga$_{1-x}$N-layers. The space group symmetry P6$_3$mc of the wurzite structure is reduced to P3m1 due to the ordering of the group III sublattice into alternating aluminium and gallium layers. The phenomenon was studied by XRD-measurements of the symmetric superlattice peaks. © 2001 Elsevier Science S.A. All rights reserved.

Keywords: AlN; RBS-channeling; Al$_x$Ga$_{1-x}$N ordering; Dislocation density

1. Introduction

The continuous alloy Al$_x$Ga$_{1-x}$N offers a wide field of applications due to a bandgap from 3.4 (GaN) to 6.1 eV (AlN) ranging from optical UV filters to insulating layers to optoelectronic devices like LED's, laser diodes or detectors [1]. A basic problem for device fabrication is the usually high dislocation density $d$ in the layers in the range of up to several $10^{10}$ cm$^{-2}$. It is shown in reference [2] that the free carrier concentration and their mobility are affected seriously by the dislocation density. A 2D epitaxial growth along monoatomic steps is suitable to obtain high crystal quality. To reduce the amount of threading dislocations (TD) homogeneously their formation process and the corresponding growth conditions have to be studied in detail. Some results are reported for the growth of GaN [3] but only little has been published for the growth of AlN. The system Al$_x$Ga$_{1-x}$N offers the possibility if bandgap engineering by changing the composition. It was recently shown that depending on the growth condition superstructures of the wurzite structure can be observed for hexagonal Al$_x$Ga$_{1-x}$N-layers [4].

2. Experimental

The Al$_x$Ga$_{1-x}$N-layers ($x = 1–0$) were grown by molecular beam epitaxy (MBE) on (0001) 6H-SiC- and (0001) $\alpha$-Al$_2$O$_3$-substrates in a Riber P32 machine equipped with gallium and aluminium effusion cells. Nitrogen radicals were produced by the OAR CARS25 RF-Plasma source with a nitrogen flow rate of 1.4 scm and a RF-power of 440 W. The substrate temperature was varied between 900 and 1050°C for the growth of AlN and 750–800°C for the growth of Al$_x$Ga$_{1-x}$N. A 350 nm AlN buffer layer was deposited prior to growth of the alloy films. Ordering in Al$_x$Ga$_{1-x}$N was analysed by measuring the intensity relation of $2\theta/2\phi$ scans of the 001 superlattice peaks and the 004 sublattice peaks using a four circle X-ray diffractometer with Cu-K$_\alpha$ radiation. The thickness and the composition of...
the layers were determined by Rutherford backscattering (RBS).

3. Results and discussion

The formation of TD was followed by TEM, RBS-channeling and by XRD-rocking curves. A transition from 2D to 3D growth was observed for very high and for low V/III-ratios resulting in an increase of FWHM of the XRD-rocking curves [5]. The observed increase corresponds to a rise of the amount of TD of more than two orders of magnitude compared to the best values of about $2 \times 10^8$ cm$^{-2}$ in the case of stoichiometric, 2D grown layers on SiC. The 2D/3D transition is traced back to a reduced surface mobility of Al on the nitrogen terminated surface. For both, SiC and sapphire substrates, cross-sectional TEM images show high dislocation densities of about several $10^{11}$ cm$^{-2}$ at the interface to the substrate of the 2D grown AlN layers (Fig. 1A). During the growth process this amount is reduced due to annihilation of the defects whose Burger’s vectors are reasonably tilted with respect to the growth direction. The amount of the tilt of Burger’s vectors depends strongly on the type of substrate and the growth conditions increased by different annihilation rates of the defects. Fig. 1B shows depth profiles of TD density obtained by RBS-channeling. In the case of 2D grown films on SiC a high annihilation rate of TD emerges close to the interface while the rate is decreased for 3D grown Al$_x$Ga$_{1-x}$N-layers and even more for layers nucleated on sapphire. But in all cases there seems to appear a second, much slower annihilation process which is attributed to TD with Burger’s vectors directed close to the growth direction. A transition from 2D step flow to 3D spiral growth is observed for substrate temperatures below 970°C. Step heights of about 0.25 nm are determined by AFM which fits the value of $c/2 = 0.258$ nm for bulk AlN [6]. Chemical ordering was observed in the XRD analysis of the Al$_x$Ga$_{1-x}$N-layers. In the ordered alloy the metal atoms occupy preferentially lattice sites with alternating of Al and Ga layers perpendicular to the [0001] growth direction. The phenomenon was observed by measuring the XRD superlattice peaks. The XRD scattering conditions for this additional peaks are $h + 2k = 3n$ and $l$ odd. Fig. 2A and Fig. 2B show the symmetric 001 superlattice and the 004 sublattice peaks of XRD $\theta$/$2\theta$

![Fig. 1. (A) (left): Cross-sectional TEM image of an AlN layer grown on SiC substrate ($g = 004$). The interface shows high dislocation density in the range of about $10^{11}$ cm$^{-2}$. Within the AlN layer the density of threading dislocations is reduced to $10^9$ cm$^{-2}$ (evaluated from TEM plan view images). (B) (right): Profiles of the dislocation density in dependence on the thickness of the epitaxial layer measured by RBS-channeling. Thickness values starting at the substrate interface.](image)

![Fig. 2. XRD $\theta$/$2\theta$ scans of the Al$_x$Ga$_{1-x}$N 001 superlattice and 004 sublattice peaks for $x = 0.5$ (left) and 0.87 (right).](image)
nihilation of defects was influenced by the type of substrate and the growth mechanism (2D-3D) due to the formation of defects with differently oriented Burgers' vectors. Higher dislocation densities were found at lower substrate temperatures. Long range ordering was observed for the continuous alloy Al$_x$Ga$_{1-x}$N. The space group symmetry P6$_3$mc of the wurzite with a random mixture of the atoms on metal sites is reduced to P3m1 due to the ordering of the group III sublattice into alternating aluminium and gallium layers. The experimental results of the XRD measurements were in good qualitative agreement with the theoretical prediction and the degree of ordering is determined by the growth conditions. The phenomenon of ordering in semiconductor alloy layers is believed to depend on the different tetrahedral radii of the atoms in the sublattices.

Acknowledgements

The authors acknowledge D. Hofmann and A. Winnacker, University of Erlangen, for supplying the SiC substrates and the government of Baden-Württemberg for funding.

References