Fundamental studies on crystallization and reaching the equilibrium shape in basic ammonothermal method

Sochacki T.[1]*, Kucharski R.[1], Grabianska K.[1], Weyher J.L.[1], Sadovyi B.[1], Iwinska M.[1], Kirste L.[2]. and Bockowski M.[1],
*lead presenter: tsochacki@unipress.waw.pl
1 Institute of High Pressure Physics Polish Academy of Sciences, Warsaw, Poland,
2 Fraunhofer Institute for Applied Solid State Physics (IAF), Freiburg, Germany

Gallium nitride (GaN) substrates of high structural perfection are needed for advanced electronic and optoelectronic devices based on GaN-on-GaN technology. The wafers can be prepared from crystals grown by three main methods: crystallization from gas phase, basic or acidic ammonothermal growth, and growth from solution of gallium and sodium. One of the most promising GaN crystallization methods is basic ammonothermal. It allows to crystalize highly conductive (n-type) and semi-insulating ammonothermal GaN (Am-GaN). Recently, 2-inch Am-GaN wafers of extraordinary structural quality have been introduced to the market. They are crystallographically flat, thus with a uniform off-cut and the threading dislocation density (TDD) does not exceed 5×10^4 cm^{-2}.

Despite many published papers devoted to crystallization as well as properties of ammonothermal crystals, a detailed investigation of basic ammonothermal growth process has never been presented. With this work we aim to fill this gap. By analyzing crystallization on a native seed of a lenticular shape, thus with a varying off-cut on its surface, we try to answer some basic questions: i/ in which crystallographic directions does the growth proceed and which crystallographic planes play the most important role?; ii/ what are the relationships between growth rates in different crystallographic directions?; iii/ what is the influence of the off-cut of the seed on the growth process? For this purpose, slices in two crystallographic directions: [-1-120] and [-1010], of a hexagonal crystal grown on a lenticular round shape seed were prepared and analyzed. Photo-etching (PEC), optical microscopy (OM) with Nomarski contrast as well as X-ray topography (XRT) and high-resolution X-ray diffraction (HRXRD) were applied as the main methods for investigating structural properties, such as striations and growth bands, dislocations, etc. Furthermore, secondary ion mass spectrometry (SIMS) was used for analyzing concentrations of impurities in all sectors of the analyzed crystal slices with different crystallographic orientations. In addition, the new-grown crystal was cut and part of the (0001) plane was subject to defect selective etching (DSE) in molten KOH-NaOH eutectic in order to analyze the etch pit density (EPD). The obtained results allowed us to create a growth model of Am-GaN crystallized on a lenticular seed. Model which illustrates the path a crystal passes to achieve an equilibrium shape [2].

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