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Self Oriented Growth of GaN Films on Molten Gallium

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The need for native or lattice matched substrates for growing single crystal quality GaN films is tremendous. The epitaxial growth on the currently available, lattice mismatched substrates produces GaN crystals with high density of misfit dislocations. These films limit the extent of usefulness of GaN as a direct, wide bandgap material for blue light emitting and laser devices. In this regard, there have been tremendous efforts to synthesize large area bulk III-nitride crystals with low defect densities. To date, bulk growth processes using high pressures and temperatures produced 15 mm sized crystals only [1]. The low-pressure synthesis demonstration experiments using atomic nitrogen over molten Ga produced only polycrystalline GaN mass [2].

We have recently demonstrated a concept in which hexagonal Gallium Nitride crystals could self-assemble during nucleation and growth on top of molten gallium to form large area self-oriented films [3]. In this paper, a systematic and thorough study is performed to understand the self-orientation within the resulting films. The direct nitridation experiments were performed using amorphous, fused- silica quartz substrates, atomic nitrogen in an ECR-MW plasma reactor and Ga films of thicknesses ranging from 2 to 500 microns. Several experiments using stationary stage showed the self-oriented regions could be as large as 5 mm or higher. The grain size estimated from top view of GaN film is about 4-5 μm with a thickness of 13 μm for nitridation time scales of 5h (Fig.1). Micro-Raman spectra showing a peak at 144 cm^{-1} confirmed the wurtzite phase of GaN. XRD θ-2θ scans showed predominant reflections of (0002) and (0004) planes indicating C-plane orientation. The cross-sectional SEM indicates the presence of molten gallium layer below the self-oriented regions (Fig.2). Cross-section TEM of the GaN shows no contrast from dislocations but indicates the presence of stacking faults (Fig.3). The misorientation between the neighboring crystals was analyzed using convergent beam electron diffraction technique. Also, several experiments were conducted using a rotating substrate stage at various speeds ranging from 30 rpm to 600 rpm. Preliminary experiments using the rotating substrate stage indicated that rotation helps to smoothen the GaN film and also helps in increasing the regions over which self-oriented GaN film occurs.

In order to understand the nucleation process, we also performed experiments using nitridation of gallium droplets with sizes ranging from 60 microns to 2 mm. The results on the estimated nucleation density and the nuclei size of the resulting GaN showed a strong correlation with the initial gallium droplet diameter. These results were reconfirmed by with several experiments at different process conditions (temperature and pressure). The results indicate that the nucleation of GaN on molten gallium takes place in the bulk and not just on the surface.

References:

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2. Angus et.al., Appl. Phys. Lett. 70, 179 (1997)
3. H. Li, H. Chandrasekaran, M. K. Sunkara, MRS Symp. Proc., V743, L3.10, (2002)

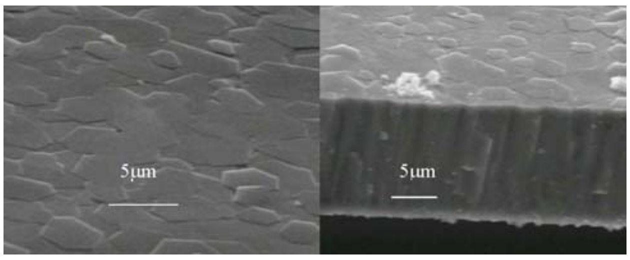

Fig.1: Self-oriented GaN flakes after Ga was dissolved away

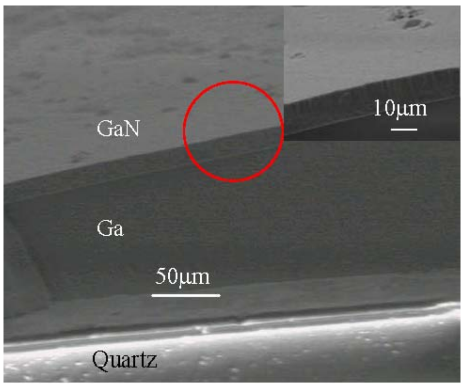

Fig.2: Self-oriented GaN on top of molten Ga

Fig.3: TEM BF image shows no dislocation contrast and some stacking faults