Detailed study of HVPE-GaN doped with silicon

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One of the most popular method for crystallizing gallium nitride (GaN) is halide vapor phase epitaxy (HVPE). It is based on reaction between gallium chloride (GaCl) with ammonia (NH_3) at 1000°C and ambient pressure. Gallium chloride is synthesized in-situ in a reactor at a lower temperature (800°C) by reacting hydrochloride (HCl) with gallium (Ga). Both foreign, sapphire or gallium arsenide (GaAs) crystals and native crystals are used as seeds. The HVPE method enables growth of GaN crystals with low level of impurities. In quartz reactor, the impurities concentrations on the (0001)-plane of unintentionally-doped (UID) HVPE-GaN grown in the <0001> direction are close to 10^{16} cm^{-3}. In turn, the free carrier concentration (n) is usually not higher than 5×10^{16} cm^{-3}. It is well known that the main impurity in UID HVPE-GaN crystallized in a quartz reactor is silicon (Si). It comes from the quartz reactor components. Oxygen (O), on the other hand, does not accumulate on the (0001) plane [1]. Silicon concentration [Si] was investigated in free-standing (F-S) UID HVPE-GaN and differences in the values on (000-1) and (0001) faces were already demonstrated [2]. According to secondary ion mass spectrometry (SIMS) measurements on the (0001) plane the [Si] was 2×10^{17} cm^{-3}, while on the (000-1) plane it was 2×10^{16} cm^{-3}. The [Si] gradient was also observed in HVPE-GaN grown with intentionally incorporated Si [3]. For a constant flow of precursor of Si, dichlorosilane (H_2SiCl_2), during a crystallization process the [Si] varied by more than one order of magnitude and was higher close to the (0001) surface. For example, the [Si] could change from 2×10^{18} cm^{-3} to 2×10^{19} cm^{-3}. Free carrier concentration gradient, studied by Raman spectroscopy (RS), was also observed, and the value was always higher near the (0001) surface. In addition, SIMS and Hall measurements performed on (0001) HVPE-GaN:Si surfaces showed that from a certain [Si] in GaN, n decreases as [Si] increases [4]. The drop of the efficiency of Si doping may be caused by the creation of Ga vacancies (V_{Ga}) and complexes of V_{Ga} with Si atoms or formation of Si-Si pair defects what was recently shown by Prohozeev et al. [5].

In order to understand and investigate the described above phenomena a few HVPE-GaN:Si crystallization processes were performed on native seeds with varying growth parameters. Morphology, structural quality and growth rate of the obtained crystals were investigated and determined. The (0001) and (000-1) surfaces as well as cross-sections (10-10)-planes, of the samples were examined. The crystals were studied by optical microscopy (OM), X-ray diffraction (XRD), SIMS, Hall measurements, and RS. The determined n was compared to the [Si]. It is shown that the cause of [Si] gradient in HVPE-GaN:Si is associated with the Si deposition inside the reactor, in the H_2SiCl_2 line, as well as with the growth morphology.

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