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Microstructure Characterization of Cu-Doped GaN Grown by Plasma Assisted Molecular Beam Epitaxy

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Searching for magnetic semiconductors with a Curie temperature above room temperature is an important challenge in current development of spintronic devices [1]. In this perspective, diluted magnetic semiconductors (DMS) based on III-nitrides alloyed with transition metal elements are of particular interest [2-4]. Among them, Cu doped GaN has attracted attention recently. First principle calculation predicted that Cu has a ferromagnetic ground state with a magnetic moment of $2\mu_B$/atom [5]. In addition, applying Cu doping can avoid controversy regarding the origin of ferromagnetic ordering since neither metallic Cu nor all possible phases in Cu-Ga-N system are ferromagnetic [6-10].

Ganz et al. [11] reported recently that Cu-alloyed GaN epilayers grown by molecular beam epitaxy (MBE) are ferromagnetic at room temperature. However, the nominal magnetic saturation is lower than theoretic prediction. This was attributed to the presence of large amount of islands enriched with Cu on the epilayers. Two important issues are raised. First, the formation mechanism of the islands has to be clarified in order to find a proper strategy to improve the Cu incorporation in GaN. Second, the real composition of Cu in GaN has to be determined to justify the magnetic moment contributed by Cu atoms.

We carried out a thorough study on the formation mechanisms of the islands formed on the Cu-alloyed GaN prepared by plasma assisted MBE. The Cu-doped GaN epilayers were prepared by plasma assisted MBE in a Ga-rich environment with Cu-to-Ga beam equivalent pressure ratio of 1.2 to 4.8 %. Islands enriched with Cu were found on all the GaN epitaxial layers as shown in Fig. 1. The large, equaxed islands, labeled A in Fig. 1, are composed of a top Cu_9Ga_4 intermetallic layer and a bottom GaN layer which is about twice as thick as the GaN layer grown from the gas phase. The thick bottom GaN layer is grown via a VLS mechanism and an orientation relationship between GaN and Cu_9Ga_4 was identified as $[111]_{Cu9Ga4}//[1\bar{2}10]_{GaN}$ and $(10\bar{1})_{Cu9Ga4}//(0001)_{GaN}$ according to the diffraction pattern shown in Fig. 2. The long, dendritic islands and the fine, equiaxed islands, labeled as B and C, respectively, in Fig. 1, are resulted from the precipitation of the Cu_9Ga_4 phase in the small, oversaturated droplets. It is reasonable to believe that Ostwald ripening plays an important role in the formation of these islands. In Ostwald ripening, atoms from the small droplets diffuse to the large ones. Due to an extremely high diffusivity of Ga on the surface, the Ga in the small droplets should be exhausted much faster than Cu and the Cu content of the droplets reached the liquidus line within a short period of time. Further outward diffusion of Ga resulted in the precipitation of the Cu_9Ga_4 phase at the AlN(GaN)/solution interface epitaxially. Wavelength dispersive X-ray spectroscopy analyses indicated that the samples contains 0.10-0.04 wt.%Cu.

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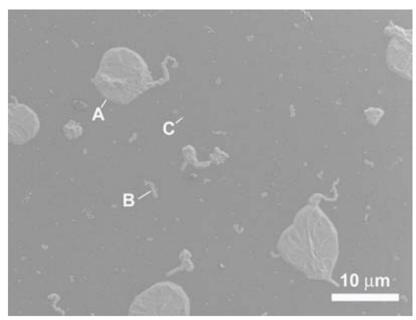

Fig. 1 Secondary electron images of the Cu-alloyed GaN having the Cu-to-Ga BEP ratio of 2.8 %.

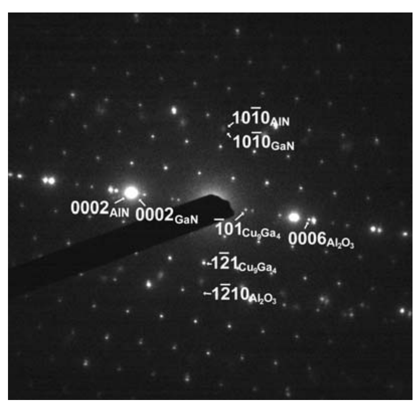

Fig. 2 Electron diffraction patterns of GaN and Cu_9Ga_4 showing a well defined orientation relationship.