

## Structural analysis and magnetic properties of nitrogen co-doped II-VI diluted magnetic semiconductor (Zn, Fe) Te thin films grown under Zn-rich condition by MBE

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In many of diluted magnetic semiconductors (DMS), the open d or f shells of the incorporated magnetic ions form a localized level within the band gap of host semiconductors [1], which opens the scope to trap carriers introduced by external impurities. Because of this trapping, the charge state of magnetic ions changes and thus makes impact on their mutual Coulomb interaction [2]. Among various II-VI DMSs, Fe-doped selenides or tellurides have been known as van-Vleck type paramagnetism [3]. The isoelectronic Fe<sup>2+</sup> state on the substitutional site of II-VI compounds produces only field-induced magnetic moments. In our previous study on nitrogen acceptor co-doped (Zn,Fe)Te thin films grown under Te-rich condition, we have observed that deviation of Fe valence state from Fe<sup>2+</sup> by N-doping has drastic impact on the magnetic properties of (Zn,Fe)Te. In this study, we have investigated the impact of co-doping of nitrogen as an acceptor impurity on the magnetic properties of (Zn,Fe)Te thin films grown under Zn-rich condition.

We grew Zn<sub>1-x</sub>Fe<sub>x</sub>Te thin films by molecular beam epitaxy (MBE) in excess of Zn flux over Te flux with the Fe composition fixed at  $x = 0.014$  and the N concentration varied in the range of  $[N] \sim 10^{18} - 10^{20} \text{ cm}^{-3}$ . We used N plasma source with N<sub>2</sub> gas excited by rf plasma. Structural analysis was done by x-ray diffraction (XRD) and x-ray absorption fine structure (XAFS) measurements. In the XRD  $\theta$ - $2\theta$  scan, no extrinsic precipitates were detected. The XAFS measurement reveals that N-doped films with intermediate N concentration  $[N] \sim 10^{18} - 10^{19} \text{ cm}^{-3}$  (#2, #3) are composed of pure diluted phase [shown in Fig. 1(a)] with the substitutional Fe atoms but their valence state deviates from Fe<sup>2+</sup> (#1, undoped film) as observed clearly from the shift of the pre-edge centroid positions in the XANES spectra [shown in the inset of Fig. 1(b)]. In the XANES spectrum, the pre-edge feature, which is located  $\sim 15 - 20 \text{ eV}$  lower than the main absorption edge of Fe, is sensitive to Fe redox states. The pre-edge position shifts towards higher energy with increasing oxidation state and the separation between the average pre-edge centroid positions for Fe<sup>2+</sup> and Fe<sup>3+</sup> is  $1.4 \pm 0.1 \text{ eV}$  [4, 5]. Magnetization measurement confirms drastic change of magnetic properties from paramagnetic (#1) to room temperature ferromagnetic (#2, #3) [shown in Fig. 1(c)], which would be attributed to the deviation of Fe valence state from Fe<sup>2+</sup> to Fe<sup>2+/3+</sup> mixed state or solely to Fe<sup>3+</sup> state due to the incorporation of N. For the N-doped film with  $[N] \sim 10^{20} \text{ cm}^{-3}$  (#4), room temperature ferromagnetism might have originated from the secondary phase of small Fe clusters.

References : [1] K. Sato and H. Katayama-Yoshida, *Semicond. Sci. Technol.* **17**, 367 (2002). [2] T. Dietl, *Nature Mater.* **5**, 673 (2006). [3] A. Twardowski, *J. Appl. Phys.* **67**, 5108 (1990). [4] L. Galois, *Chem. Geo.* **174**, 307 - 319 (2001). [5] M. Wilke, *Am. Mineral., J. Ear. Plan. Mater.* **86**, 714 - 730 (2001).

