ROBL-CRG	**Experiment title:** **Nanophase formation in ZnO implanted with transition or rare earth metal ions**	**Experiment number**: 20-02-634
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Report:

Recently due to the potential application in spintronics, both magnetic/semiconductor hybrid heterostructure and diluted magnetic semiconductor (DMS) have attracted huge research attention [1]. In both research fields, GaAs is the most intensively studied material. Epitaxial MnAs and transition metal (e.g. Fe) were achieved on GaAs substrates due to their structure compatibility: i.e. Mn-based metallic compounds have common III/column atoms with GaAs, while Fe/GaAs has a cube-on-cube orientation with a small lattice mismatch of -1.4% [2, 3]. Room temperature spin injection was reported from MnAs [4] and Fe [5] into GaAs. In parallel, Mn doped GaAs has been demonstrated to be a DMS with a Curie temperature of 110 K [6]. Very recently, wide-band-gap semiconductors (GaN and ZnO) doped with transition metals were reported to be DMS with Curie temperatures above room temperature [7]. Nevertheless, the origin of the observed ferromagnetism is still controversial, e.g. ferromagnetic clusters [8]. In contrast, only a few investigations deal with epitaxial magnetic/ZnO(or GaN) heterostructures. This is partially due to the chemical incompatibility, the different crystal symmetry or the large lattice mismatch between the 3d-ferromagnet and ZnO. For instance, Fe is a bcc crystal, while hcp MnAs and Co have a very large lattice mismatch with ZnO (12% and 23%, respectively). In this paper, we made an extensive investigation on the structural and magnetic properties evolution of Fe implanted ZnO upon annealing, and demonstrate the possibility to form epitaxial magnetic Zn-ferrite embedded in ZnO. With respect to the crystal symmetry and lattice mismatch, our results suggest that other ferrites, which have been epitaxially grown onto MgO, $SrTiO_3$, and $Y_{0.15}Zr_{0.85}O_2$, and exhibit rich magnetic properties [9], could be epitaxially embedded inside or grown onto ZnO.

From the experiment we have reached two conclusions:

(1) By correlating the structural and magnetic properties of Fe-implanted ZnO, we found either bcc-Fe or partially inverted $ZnFe_2O_4$ nanocrystals are the origin of the observed ferromagnetism.

(2) We demonstrate the possibility to form epitaxial magnetic Zn-ferrite embedded in ZnO, and the epitaxial relationship is $ZnFe_2O_4$ (111)[110]//ZnO(0001)[11$\underline{2}$0].

Fig. 1 shows the SR-XRD patterns for the as-implanted and annealed samples. For the as-implanted sample, Fe nanoparticles were observed, and no other Fe-oxide (Fe_2O_3, Fe_3O_4, and $ZnFe_2O_4$) particles were detected. After 823 K and 15 min annealing, larger and more Fe nanoparticles are formed. After 1073 K and 15 min annealing, the Fe(110) peak almost disappeared and the sample already shows an indication for the presence of $ZnFe_2O_4$. After 3.5 hours annealing at 1073 K, crystalline and oriented $ZnFe_2O_4$ particles are clearly identified. The inset shows a zoom of the Fe(110) peak to show the development of Fe nanoparticles more clearly.

Fig. 2 shows the pole figure for $ZnFe_2O_4$(511) and $ZnFe_2O_4$(333). Both diffraction lines have the same Bragg angle in the cubic $ZnFe_2O_4$. The sample tilt, χ, is the angle by which the surface is tilted out of the diffraction plane. The angle of rotation about the surface normal is denoted by φ, which ranges from -60° to 60°. The pole figure shows poles at χ~39°, 56° and 70°, respectively, with sixfold symmetry and an indication of a tail from ZnO(1120). It is consistent with the theoretic $ZnFe_2O_4$(511) / (333) pole figure viewed along [111] with rotation twins. The in-plane orientation relationship is $ZnFe_2O_4$[110]//ZnO[1120].

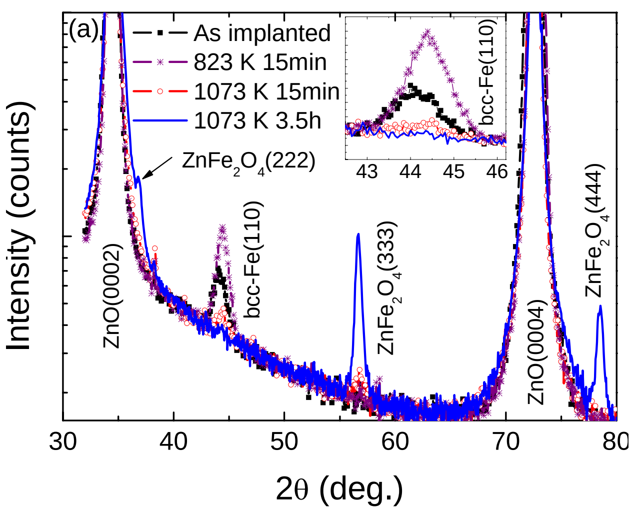

Fig. 1: SR-XRD patterns of Fe implanted ZnO reveal the second phase development (from bcc-Fe to $ZnFe_2O_4$) upon annealing.

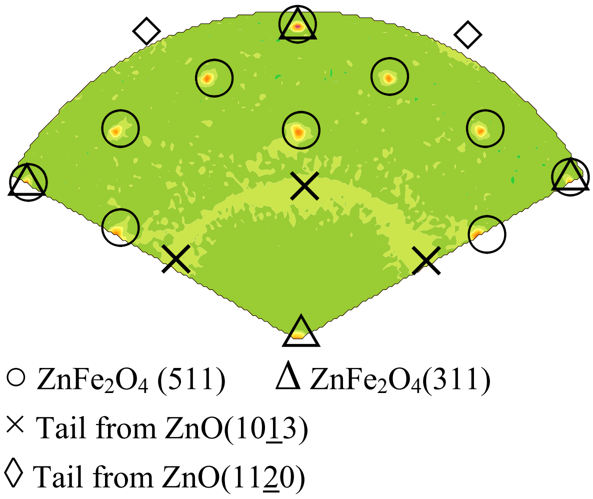

Fig. 2: Pole figure of $ZnFe_2O_4$(511) reveals the epitaxy of $ZnFe_2O_4$.

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