Interpretation of ferromagnetic Fe doped ZnO by Mössbauer spectroscopy

Seung-Iel Park, Geun Young Ahn, and Chul Sung Kim

Department of Physics, Kookmin University, Seoul 136-702, Korea

(Submitted on 11 January 2007; received 31 October 2006; accepted 23 December 2006; published online 3 May 2007)

Single phase Zn$_{0.95}$Fe$_{0.05}$O sample was obtained by the sol-gel method with annealing at 650 °C for 6 h in H$_2$ 5%/Ar balance gas atmosphere. The crystalline structure of Zn$_{0.95}$Fe$_{0.05}$O is determined to be a P6$_{3}$mc hexagonal structure with lattice constants $a_0=3.255$ Å and $c_0=5.207$ Å at room temperature. The Mössbauer spectra were obtained at various temperatures ranging from 4.2 to 295 K. The values of the isomer shifts ($\delta$) show that for all temperature ranges, they are in the ferrous (Fe$^{2+}$) state. The magnetic hyperfine field ($H_{hf}$) and electric quadrupole splitting ($\Delta E_Q$) in the weak ferromagnetic state at 4.2 K have been analyzed, yielding the following results: $H_{hf} = 37.8$ kOe, $\theta = 67.5^\circ$, $\varphi = 0^\circ$, $\eta = 0.75$, $\Delta E_Q = 2.06$ mm/s, and $R = 7.4$, respectively. From the Mössbauer spectrum at 77 K, the paramagnetic quadrupole phase is related to the temperature dependence of spin-lattice relaxation. © 2007 American Institute of Physics.

[DOI: 10.1063/1.2712527]

I. INTRODUCTION

Transition metal (Fe, Co, Ni, etc.) doped ZnO based diluted magnetic semiconductors have been studied by many researchers since Dietl et al. demonstrated ferromagnetic properties at room temperature by dilute doping with a transition metal. Recently, the ferromagnetic properties for the ZnO based magnetic semiconductor at room temperature was elucidated to occur with the oxygen defects in zinc oxide and its various charge states are due to the covalent oxygen-oxygen bond. Also, researches on TiO$_2$ based magnetic semiconductor have taken this influence into consideration.

In this paper, we present the results of Mössbauer experiments for the $^{57}$Fe doped ZnO sample which were analyzed with the consideration of $H_{hf}$, $\omega e^2 Q$, $\theta$, $\varphi$, and $\eta$ and compare them with those of the x-ray diffraction analysis.

II. EXPERIMENT

Single phase polycrystalline Zn$_{0.95}$Fe$_{0.05}$O was obtained by the sol-gel method with annealing in hydrogen/argon. The crystalline structure of the sample was analyzed by the long time scanning of 2 h using a Philips x-ray diffractometer with Cu K$\alpha$ radiation. For the analysis with the hyperfine magnetic property, Mössbauer spectra were measured at various temperatures ranging from 4.2 to 295 K.

III. RESULTS AND DISCUSSION

Figure 1 shows the x-ray diffraction patterns of the Zn$_{0.95}$Fe$_{0.05}$O sample at room temperature by Rietveld refinement analysis. The crystalline structure of Zn$_{0.95}$Fe$_{0.05}$O was determined to be a wurtzite hexagonal symmetry of P6$_{3}$mc with lattice constants $a_0=3.255$ Å and $c_0=5.207$ Å at room temperature, while the Bragg $R_B$ and $R_f$ factors were 2.60% and 2.05%, respectively. The composition of $^{57}$Fe doped ZnO sample was determined to be Zn$_{0.95}$Fe$_{0.05}$O$_{1.21}$ by x-ray diffraction analysis. These defects in the Fe doped ZnO sample are due to the annealing condition in the H$_2$ 5%/Ar balance gas atmosphere. With the x-ray analysis results, we considered a highly symmetric state for interstitial oxygen, which is identical to the investigation of Erhart et al. and Zhang co-workers. This phenomenon is responsible for the ferromagnetic interaction in the ZnO structure.

Figure 2 shows the applied field dependence of the magnetic curve at 60 K and room temperature. For the substitution of isotope $^{57}$Fe, the ferromagnetic property appeared to be smaller when compared with the natural Fe substituted. However, as shown in Fig. 2, a shape of very weak ferromagnetic behavior is shown at room temperature. This result was explained with a highly symmetric state for interstitial oxygen bond.

FIG. 1. The x-ray diffraction pattern of the Zn$_{0.95}$Fe$_{0.05}$O sample at room temperature. The open circles represent the observed patterns; continuous lines represent calculated and difference (obs-cal) patterns. The tick marks correspond to the position of the allowed Bragg reflections.

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$^{a}$Present address: Department of Neutron Physics, HANARO, KAERI, Yuseong, Daejeon 305-600, Korea.

$^{b}$Author to whom correspondence should be addressed; FAX: +82-2-910-5170; electronic mail: cskim@kookmin.ac.kr

JOURNAL OF APPLIED PHYSICS 101, 09H113 (2007)