Materials Science

16A/2008G010

Effects of hole doping in the ferromagnetic semiconductor Mn-doped ZnO thin film studied by x-ray magnetic circular dichroism

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Introduction

Diluted magnetic semiconductors (DMSs), where magnetic ions are doped into semiconductor hosts, continue to attract considerable attention due to the possibility of utilizing both the charge and spin degrees of freedom in the same materials, allowing us to design a new generation spin electronic devices with enhanced functionalities [1]. First-principles calculations by Sato and Katayama-Yoshida [2] have predicted that ZnO-based DMSs exhibit ferromagnetism, and room temperature ferromagnetism in Mn-doped ZnO (ZnO:Mn) thin films has indeed been reported [3]. Although there is evidence showing that Mn substitutes for Zn in a 2+ state [3] and the carrier-induced mechanism for ferromagnetisn has been proposed to predominate [4], the origin of the ferromagnetic interaction in the ZnO:Mn system remains to be clarified. Thus, to achieve better understanding of this type of DMSs and to perform new material design, investigation of the electronic structure of the ZnO:Mn system is crucial.

In this report, we report on soft x-ray absorption spectroscopy (XAS) and magnetic circular dichroism (XMCD) studies of the ZnO:Mn thin film, which is hole-doped through N_2 treatment and exhibits room temperature ferromagnetism [4].

Experimental condition

A 400-nm-thick thin film of ZnO:Mn (Mn = 2 %) was grown on Si substrate by reactive sputtering using 99.9 % pure Zn target using the radio frequency (RF)/direct current (DC) sputtering system. In order to dope the system with holes, the ZnO-DMS was prepared under nitrogen pressure of P_N^2 = 4.0×10^{-5} mbar. XMCD measurements were performed at BL-16A. The monochromator resolution was E/ΔE > 10000 and the circular polarization of x-rays was higher ~ 95 %.

Results and Discussion

Figure 1(a) shows the Mn 2*p*-3*d* XAS spectra of the ZnO:Mn sample for opposite-magnetization directions recorded using circular polarized x-rays and their difference spectrum, i.e., XMCD spectrum [shown in Fig. 1(b)]. Here, the XAS spectra obtained at H ~ +5.0 and -5.0 T are denoted by σ^+ and σ^-, respectively. The XAS and XMCD spectra show multiplet structures reflecting the localized nature of the Mn 3*d* electrons. Figure 1(c) shows the XMCD spectrum of the ZnO:Mn in comparison with those of Mn-doped GaAs [GaAs:Mn, Mn^{2+} (T_d)] (Ref. 5), $CaMn_{1-x}Ru_xO_3$ [CMRO, Mn^{3+} and Mn^{4+} (O_h)] (Ref. 6) and Mn metal (Ref.7). The spectral line shape of the ZnO:Mn is nearly identical to that of the GaAs:Mn, indicating that the magnetically active component of ZnO:Mn is mainly the Mn^{2+} ions tetrahedrally coordinated by high-spin state [see the inset of Fig. 1(b)].

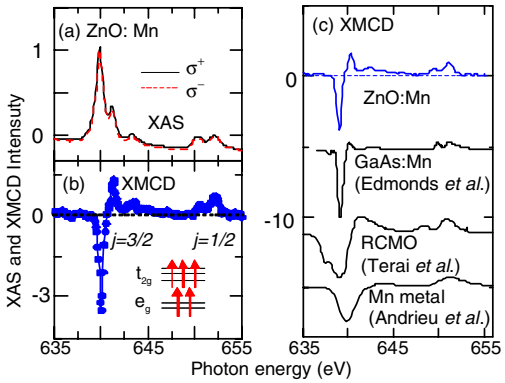

Fig. 1. Mn 2*p*-3*d* XAS (a) and XMCD (b) spectra of the ZnO:Mn. Inset picture in (b) shows the spin configuration of the Mn^{2+} ion coordinated tetrahedrally with high-spin state. (c) XMCD spectrum of the ZnO:Mn compared with those of (GaAs:Mn) [5], $CaMn_{1-x}Ru_xO_3$ (CMRO) [6] and Mn metal [7].

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