Chemical Preparation and Characterization of Transparent Ferromagnetic Fe:ZnO Semiconductor Films

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Magnetic semiconductors have been attracted much attention as a key material for spin dependent devices (spintronics devices) using optical-magnetic-electrical correlation effects.[1-3] Moreover, ferromagnets with a high optical transparency in visible light region are a novel material and are expected as a device with enhanced magneto-optical effects.[4] Ferromagnetism with Curie temperature enough higher than room temperature and low fabrication costs are required to employ these materials to commonly used devices. Although physical deposition techniques such as a molecular beam epitaxy (MBE) are generally used to prepare magnetic semiconductors, there are some issues on aspects of its mass productivity and costs. In the present study, we investigated a chemical solution process to prepare a transparent ferromagnetic Fe-doped ZnO semiconductor film and its structural, optical, electrical, and magnetic properties.

The Fe-doped ZnO films ($Fe_xZn_{1-x}O$, $x \approx 0.19$) with 250 nm in thickness were prepared by a simple 2-step procedure composed of (i) chemical deposition of <0001> oriented ZnO film on a catalyzed glass from an aqueous solution[5,6] and (ii) chemical introduction of Fe ions in another aqueous solution to the ZnO film.[7] These solutions contain only the corresponding nitrate salt and dimethyl amineborane (DMAB) and were operated at temperatures of 50-60 °C.

The as-deposited films were analyzed by the following measurements: XRD, UV-VIS photometry, Hall-effects, and VSM. The film showed room temperature ferromagnetism with 7.0 emu cm^{-3} in saturation magnetization and 60 Oe in coercive force and high transparency in visible light regions as well as *n*-type semiconducting feature with 3.6 $cm^2\ V^{-1}\ s^{-1}$ in mobility and 1.0×10^{10} cm^{-3} in carrier concentration. In the XRD pattern for the film, no reflections assigned as iron compounds could be observed and the film was identified as <0001> oriented ZnO with wurtzite structure. Differences in peak position and FWHM of 0002 reflection could not be recognized among the ZnO and Fe:ZnO films.

In order to clarify the origin of room temperature ferromagnetism of the Fe:ZnO film, we measured x-ray absorption fine structure (XAFS) and soft x-ray magnetic circular dichroism (SXMCD) at Fe and Zn *K*, $L_{2,3}$ edges in Japan Synchrotron Radiation Institute (JASRI), SPring-8. X-ray absorption near-edge structure (XANES) indicated that ferric and ferrous ions are contained in the film, and k^3 weighted EXAFS oscillation $k^3\chi(k)$, where k is wave number, revealed that the local structure around the introduced iron was analogous to that of bulk ferromagnetic magnetite (Fe_3O_4). In an SXMCD measurement on Fe $L_{2,3}$ edges, MCD was observed as shown in Figure 1. The MCD spectrum for the Fe:ZnO film was almost the same in profile and peak energies as those for ferrites with a spinel structure containing ferric ions on the tetrahedral and octahedral sites and ferrous ions on the octahedral sites. The peak intensity ratio was slightly different to those for stoichiometric Fe_3O_4, suggesting that the Fe:ZnO film contained non-stoichiometric Fe_3O_4 to some extent.

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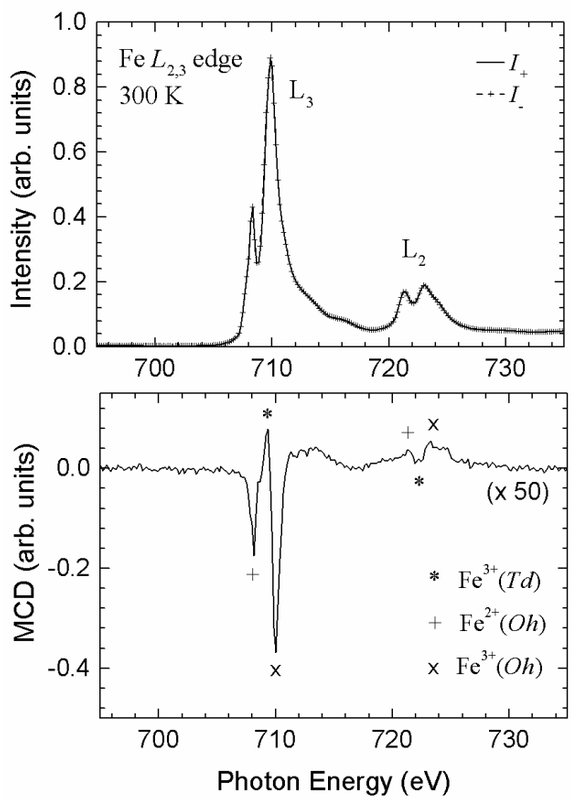

Figure 1. Normalized Fe $L_{2,3}$ XAS (top) and MCD (bottom) spectra for the $Fe_{0.19}Zn_{0.81}O$ film at 300 K. The curves I_+ (I_-) represent the XAS intensity for the right (left) circularly polarized light, respectively.