Resonant Inelastic X-Ray Scattering Study of Diluted Magnetic Semiconductor (In,Fe)As

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Diluted magnetic semiconductor (DMS) is a semiconductor doped with magnetic impurities such as 3*d* transition metal of several percentages. DMSs showing ferromagnetism have attracted much attention in a field of *spintronics* because of the ferromagnetic properties related to the carrier concentration, so-called *carrier-induced ferromagnetism*, which enables us to manipulate both charge and spin degrees of freedom of the carrier in the semiconductor [1]. An archetypical DMS $Ga_{1-x}Mn_xAs$ shows *p*-type carrier-induced ferromagnetism [2]. Indeed, using $Ga_{1-x}Mn_xAs$, novel functional spintornic devices have been fabricated so far even though the Curie temperature is below room temperature [3, 4]. To expand the applications of DMS to spintronic devices, *n*-type ferromagnetic DMSs are desirable. Recently, Hai *et al*. [5, 6] have succeeded in creating a new *n*-type carrier-induced ferromagnetic DMS $In_{1-x}Fe_xAs$:Be. Here, Be ions are located on the interstitial site due to the low-temperature growth condition and act as double donor. One can independently control the concentrations of magnetic impurity and electron carrier in $In_{1-x}Fe_xAs$:Be by changing the Fe and Be contents. When the electron carrier concentration is larger than $10^{19}cm^{-3}$, $In_{1-x}Fe_xAs$:Be shows ferromagnetic property. The magnetization curves measured by SQUID are identical to those by anomalous Hall effect and magnetic circular dichroism (MCD) in visible-ultraviolet reflection [5], indicating that the ferromagnetic property in $In_{1-x}Fe_xAs$:Be is derived from an intrinsic carrier-induced ferromagnetism. However, mechanism of the ferromagnetism in $In_{1-x}Fe_xAs$:Be still remains under dispute. Knowledge of the electronic structure of the Fe ion is indispensable for revealing the mechanism of the ferromagnetism. In this study, we have conducted resonant inelastic x-ray scattering (RIXS) on $In_{1-x}Fe_xAs$:Be.

$In_{1-x}Fe_xAs$:Be (x = 0.05) thin films with the thickness of 20 nm were grown on InAs substrates by a molecular beam epitaxy method. To avoid surface oxidation, the films were covered by As capping layers after the deposition of $In_{1-x}Fe_xAs$:Be layers [7]. The Curie temperatures of the samples were ~30 K, as determined by Arrott plot of the MCD. The RIXS and x-ray absorption spectroscopy (XAS) experiments were performed using the high-resolution soft x-ray emission station HORNET [8] at the long undulator beamline BL07LSU of SPring-8, Japan [9]. The total energy resolution for the RIXS experiments was about 280 meV at the Fe L_3 edge. The RIXS spectra were measured with linear horizontal polarization at room temperature under a vacuum of 1.0×10^{-5} Pa. The XAS signals were collected in the total-fluorescence-yield (TFY) and total-electron-yield (TEY) modes.

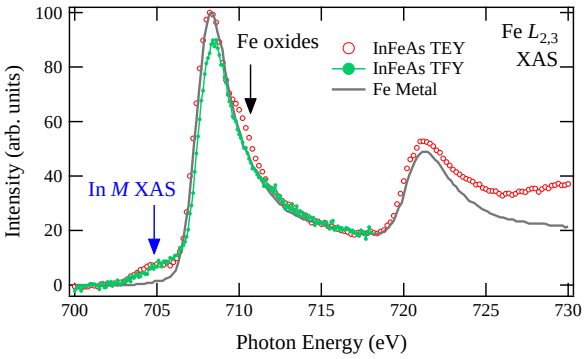

FIG.1: Fe $L_{2,3}$ XAS spectra of the $In_{1-x}Fe_xAs$:Be thin films. The signals were collected in the TEY and TFY modes. The XAS spectrum of Fe metal [11] is also shown as a reference.

Figure 1 shows Fe $L_{2,3}$ XAS spectra of the

$In_{1-x}Fe_xAs$:Be thin films taken with the TEY and TFY modes. Noted that line shape of the XAS spectra shows single-peak, broad features similar to those of Fe metal [10] and Fe pnictides [11, 12]. Therefore, correlation between the Fe 3*d* electrons in $In_{1-x}Fe_xAs$:Be is expected to be weaker as in the case of Fe pnictides compared with, for example, that in transition-metal oxides. Since the shoulder structure around 710 eV is only found in the spectrum taken in the TEY mode, the structure shall be originated from extrinsic oxidized Fe located on the surface or capping overlayer.

Figure 2 shows the Fe $L_{2,3}$ RIXS spectra measured by various excitation energies. The small Raman component, whose photon energy changes linearly with the excitation energy, is broader than the energy resolution. In $Ga_{1-x}Mn_xAs$, the Mn L_3 RIXS spectra show broadened *d-d* excitation peaks [11]. Taking into account the Mn electronic structure of $Ga_{1-x}Mn_xAs$ dominated by charge transferred states through the hybridization between Mn 3*d* and ligand, the broadening can be attributed to the lifetime broadening in the final states of the RIXS process where fast decay of the *d-d* excitations to an electron-hole pair in the host valence and conduction bands [11]. Since the electronic structure of the Fe in $In_{1-x}Fe_xAs$:Be will be similar to that of Fe pnictide, in which the Fe 3*d* orbital is well hybridized with the ligand As 4*p* band, the broadening of the Fe L_3 Raman component can be explained by the same RIXS process as $Ga_{1-x}Mn_xAs$. The Fe L_3 fluorescence component, which has the specific photon energy independent of the excitation energy, is dominant in the RIXS spectra, implying the weak electron correlation between the Fe 3*d* electrons in $In_{1-x}Fe_xAs$:Be.

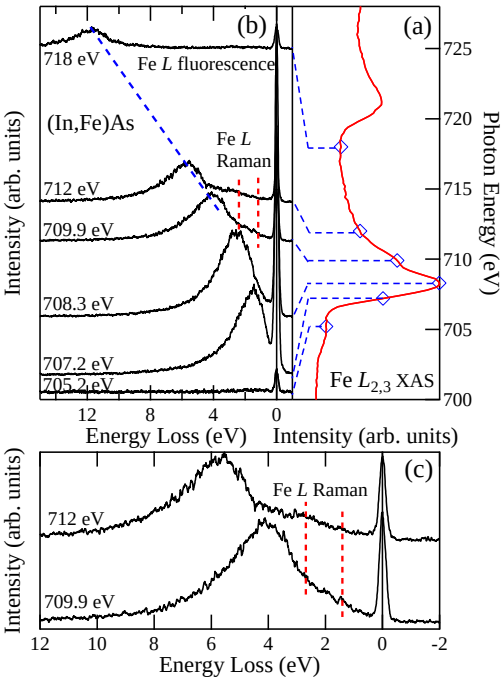

FIG.2: Fe $L_{2,3}$ RIXS spectra of the $In_{1-x}Fe_xAs$:Be thin films. (a) Fe $L_{2,3}$ XAS spectrum. Rhombuses denote the excitation energies for RIXS. (b) Fe $L_{2,3}$ RIXS spectra. (c) Enlarged plot around the Raman components.

In conclusion, we have conducted Fe $L_{2,3}$ XAS and RIXS measurements on ferromagnetic semiconductor $In_{1-x}Fe_xAs$:Be, and found the electronic structure of the Fe is well hybridized with the ligand bands similar to Fe pnictides. The observations indicate the weak electron correlation between the Fe 3*d* electrons. Following these arguments, it is concluded that the mean-field picture [2] is appropriate to understand the mechanism of the ferromagnetism in $In_{1-x}Fe_xAs$:Be, in contrast to the 3*d*-derived impurity band picture in $Ga_{1-x}Mn_xAs$. Further electronic structure studies including the valence band structure will be necessary to reveal the detailed mechanism of the ferromagnetism.

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