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Magnetization Process of the *n*-type Ferromagnetic Semiconductor (In,Fe)As:Be Studied by X-ray Magnetic Circular Dichroism

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1 Introduction

(In,Fe)As:Be is a new *n*-type ferromagnetic III-V semiconductor, in which by doping magnetic Fe atoms and Be double-donor atoms into InAs, *n*-type carrier-induced ferromagnetism is realized [1-3]. In a previous study, ferromagnetic domains with locally high electron concentration whose size was ~10 μm were observed by microscopic magneto optical imaging, explaining the hysteretic behavior of magnetization [2]. In order to investigate the nature of the magnetism on the nanometer scale, we have performed x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) measurements on (In,Fe)As:Be samples doped with 5% Fe and 10% Fe. Recently, an XMCD study of 5% Fe sample has revealed that Fe has a substantial orbital magnetic moment and that the hysteretic behavior exists at the Fe $L_{2,3}$ edges [4].

In XMCD measurements, by utilizing the absorption edges of atoms, one can access element specific information excluding extrinsic effects such as magnetic contaminations or diamagnetic response of the substrate, which overlaps the intrinsic paramagnetic component. In addition, XMCD sum rules make it possible to obtain orbital magnetic moment and spin magnetic moment separately [5, 6].

2 Experiment

The samples were fabricated using the low temperature molecular beam epitaxy (LT-MBE) method and had the structure of amorphous As cap (a few mono layer) /InFeAs:Be (20 or 30 nm)/InAs:Be buffer layer (50 nm) /InAs substrate. We have studied two different samples $In_{0.95}Fe_{0.05}As$:Be and $In_{0.9}Fe_{0.1}As$:Be. The amount of Be was estimated to be 2.6 x 10^{-19} cm^{-3} for both samples from the growth condition. Careful sample characterization by transmission electron microscopy (TEM) and energy dispersive X-ray spectroscopy (EDX) proved that there are no Fe precipitates or secondary phases in the samples. The distribution of Fe atoms was found to be uniform as long as EDX could resolve. Experiment was done at the variable-polarization undulator beamline BL-16A2 of Photon Factory (PF). Temperature was varied from 20 K to 260 K and magnetic field from 0 T to 5 T. The total electron yield (TEY) mode was used. The direction of the incident x-rays and magnetic field was perpendicular to the sample surfaces.

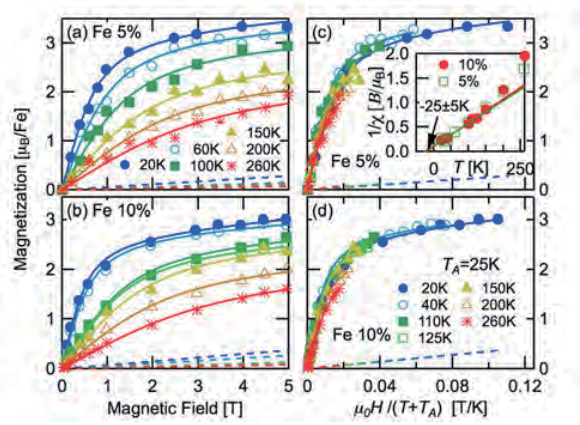

Fig. 1 (a), (b) Magnetic field dependent magnetization at various temperatures. (c), (d) Scaled plot of magnetization versus $\mu_0 H/(T + T_A)$, where T_A is analogous to antiferromagnetic Weiss temperature and is introduced so that the data at low temperatures superpose. As shown in the inset, T_A is estimated to be 25 K by linearly extrapolating the temperature dependent inverse susceptibility data to low temperature.

3 Results and Discussion

The line shapes of the XAS and XMCD spectra resemble those of Fe metal at first glance, but the peak position of XAS and that of XMCD do not coincide with each other, which is distinct from Fe metal. In addition, the ratio of the orbital moment to the spin moment estimated using XMCD sum rules was significantly larger than that of Fe metal. These facts rule out the possibility of Fe metal precipitation in these films [4].

Magnetization per Fe atom at various magnetic fields and temperatures deduced from the XMCD intensity are plotted in Figs. 1(a) and (b). In the present study, hysteresis could not be clearly observed unlike in the previous study [4] because it was below the detection limit of the present experimental set-up. At low magnetic fields, a steep increase of magnetization was observed and, at high magnetic fields, linear increase of

magnetization was observed even at low temperature of 20 K, where ferromagnetic component would saturates. This behavior can be attributed to superparamagnetism (SPM) of nano-scale ferromagnetic clusters (FC) and paramagnetism, respectively, and described by the following function,

$$M = xm_{\mathrm{Fe}}\,\mathrm{L}\left(\frac{\mu_{FC}\mu_0 H}{k_{\mathrm{B}}(T+T_A)}\right)+(1-x)\frac{C}{T+T_A}B,$$

$$\mathrm{L}(\xi) := \coth(\xi)-\frac{1}{\xi},$$

where x denotes the ratio of Fe atoms participating in the SPM or ferromagnetism to the total number of Fe atoms, m_{Fe} the average magnetic moment of ferromagnetic Fe atom, μ_{FC} the total magnetic moment per FC, T_A the antiferromagnetic "Weiss temperature", C the Curie constant of the Fe^{3+} ion with $5\mu_{\mathrm{B}}$, and L(ξ) is Langevin function. The temperature dependent susceptibility are shown in the inset of Fig. 1(c) where the linear relation could be seen at low temperatures and the T_A was estimated to be -25 K ± 5 K by extrapolation to below T = 0 K. This value of -25 K is comparable to the Weiss temperature (= -32 K) of paramagnetic (Ga,Fe)As without any co-doping or carriers doping [7]. The deviation from the linear relation at higher temperature indicates the gradual disappearance of ferromagnetism. With this antiferromagnetic T_A, the magnetization plotted as a function of $\mu_0 H/(T + T_A)$ falls on to one curve as shown in Figs. 1(c) and 1(d), and the M-H data were well fitted to the function written above. This indicates that the microscopic ferromagnetic domains consist of nano-scale FCs whose crystal structure is zinc-blende. The results of the fitting are depicted in Fig. 1 by solid curves, and the paramagnetic component is separately shown by dashed lines. The fitted parameters are shown in Figs. 2 (a) and (b). Note that fitted m_{Fe} sometimes exceeded $5\mu_{\mathrm{B}}$ if it was treated as a free parameter, and therefore, we have fixed the parameter to be $5\mu_{\mathrm{B}}$, the ideal value of high spin Fe^{3+}. The ratio of the number of ferromagnetic Fe atoms to the total number of Fe atoms was estimated to be 40%-70% and decreased as temperature increased. The magnetic moment per SPM cluster was 300-400μ_{B} for both samples, which corresponds to 60-80 Fe atoms in one cluster, and they remained nearly constant or slightly decreased as the temperature increased. The mechanism of how these FCs are formed remains unclear at present, but the local nanometer-scale variation of electron or Be density is a possible origin that weakly bound electrons around Be impurities stabilize the ferromagnetism, which is often discussed in terms of bound magnetic polarons (BMPs).

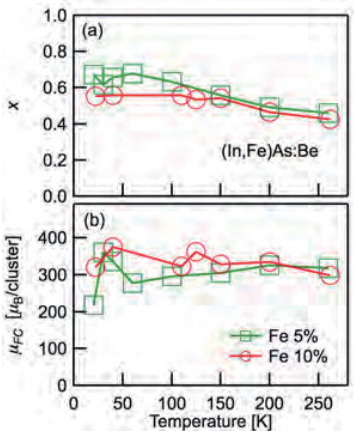

Fig. 2 (a), (b) Fitted parameters. x : ratio of Fe atoms participating in ferromagnetism to all Fe atoms. μ_{FC} : magnetic moment per ferromagnetic cluster (FC).

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