Nature of Magnetic Coupling between Mn Ions in As-Grown $Ga_{1-x}Mn_xAs$ – Systematic Temperature and Magnetic Field Dependent XMCD –

$Ga_{1-x}Mn_xAs$ is the prototypical and most well-characterized diluted magnetic semiconductor (DMS) [1]. Although many studies have been performed intensively to obtain a high Curie temperature (T_C) exceeding room temperature, it has not been achieved yet. Because $Ga_{1-x}Mn_xAs$ is grown under thermal non-equilibrium conditions, it is difficult to avoid the formation of various kinds of defect and/or disorder, which might lead to any non-ferromagnetic component. The characterization of the non-ferromagnetic Mn ions is therefore a clue to identifying how they are related to ferromagnetic ordering and eventually to improving the ferromagnetic properties of $Ga_{1-x}Mn_xAs$ samples. However, it has been difficult to extract the above information through conventional magnetization measurement owing to the large diamagnetic response of the substrate and the unavoidable mixture of magnetic impurities. We have performed an X-ray magnetic circular dichroism (XMCD) measurement to address the problem.

X-ray magnetic circular dichroism (XMCD) is defined as the difference between X-ray absorption spectra (XAS) intensities of a ferromagnetic material for circularly polarized photons that are parallel and antiparallel to the orientation of the magnetization of the material. One can investigate and/or characterize the magnetic properties of a specific magnetic element. In the soft X-ray region (400 eV - 2000 eV), especially, the information on the spin electronic structures can be obtained directly because the $L_{2,3}$ absorption edge of transition metals ($2p$ - $3d$) exists. Using the sum rules for the XAS and XMCD signal [2], the spin and orbital magnetic moments of a specific element can also be estimated quantitatively.

In the present study, in order to characterize the magnetic behaviors of substitutional Mn (Mn_{sub}) and interstitial Mn (Mn_{int}), we performed systematic T (sample temperature)- and H (magnetic field)-dependent XMCD studies in the Mn $L_{2,3}$ absorption edge region of $Ga_{1-x}Mn_xAs$. The experiments were performed at beamline **BL23SU** using the XMCD apparatus shown in Fig. 1. T and H can be scanned from < 6 K to room temperature and from 0 to 10 T, respectively. Recently, we have improved the XMCD measurement system for the effective measurement for a short time by installing a nonstop measurement system [3]. Using the nonstop measurement system, we can perform systematic T- and H-dependent XMCD measurements under many experimental conditions. We prepared two as-grown samples with different Mn concentrations, $x = 0.042$ and 0.078, whose T_C values were ~60 and 40 K, respectively. The XAS were obtained by the total electron yield mode. The measurements were carried out without surface treatment and H was applied to the sample perpendicular to the film surface.

Figure 2 shows the XAS (μ^+ and μ^-) in the photon energy region of the Mn L_3 absorption edge and the

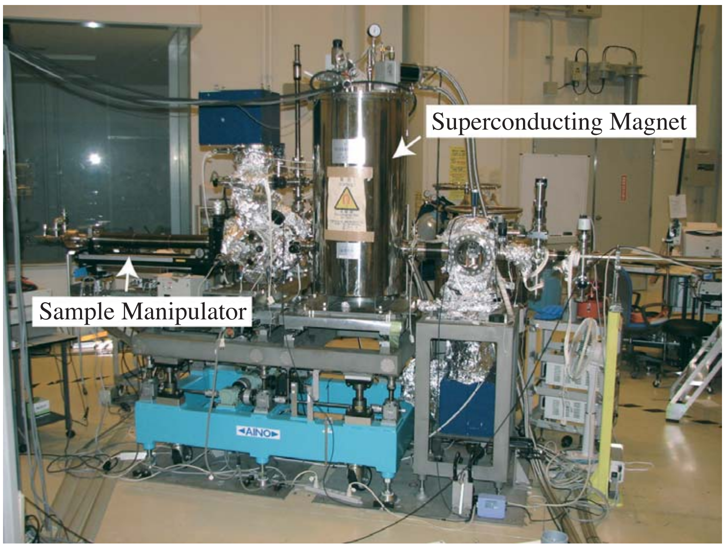

Fig. 1. XMCD apparatus of the soft X-ray beamline BL23SU.

corresponding XMCD spectra, defined as (μ^+ - μ^-), at T = 20 K and H = 0.5 T for x = 0.078. Here, μ^+ (μ^-) refers to the absorption coefficient for the photon helicity parallel (anti-parallel) to the Mn 3d majority spin direction. The magnitude of XMCD signals shows significant T and H dependences. Using these XAS and XMCD spectra, we applied the XMCD sum rules [2]. The spin magnetic moments (M_S) at T=20 K and H=0.5 T are estimated to be M_S=2.5±0.2 and 1.7±0.2 (μ_B per Mn) for x=0.042 and 0.078, respectively.

Figure 3 shows the H dependence of M_S at several temperatures for x=0.042 [panel (a)] and 0.078 [panel (b)] obtained by the XMCD measurements. Above 0.5 T, M_S increases almost linearly as a function of H. At T=20 K, the slope (dashed lines) and $M_S|_{H\to 0T}$ are smaller for x=0.078 than for x=0.042, suggesting that the anti-ferromagnetic (AF) interaction becomes stronger for x=0.078 than for x=0.042. This is reasonable because the number of Mn_{int} is expected to be larger for larger Mn concentrations. Assuming that the M_S per Mn_{sub} is 5 (μ_B per Mn) and the M_S of Mn_{int} is antiparallel to that of Mn_{sub}, the ratio of Mn_{int} atoms in the intrinsic component (R_{int}) is estimated to be 0.26 for x=0.042 and 0.33 for x=0.078 from $M_S|_{H\to 0T}$ at 20 K. Therefore, this means that the number of Mn_{int} ions should be strongly related to T_C, namely, Mn_{int} ions suppress the ferromagnetic properties by ordered spins of Mn_{sub} ions.

In conclusion, we have investigated the T, H and Mn concentration dependences of the ferromagnetism in as-grown $Ga_{1-x}Mn_xAs$ samples by XMCD measurements. The present results indicate that the AF interaction between Mn_{sub} and Mn_{int} ions, which is enhanced as the Mn concentration x increases, plays an important role in determining the magnetic behavior of $Ga_{1-x}Mn_xAs$. In addition, the number of Mn_{int} ions should be strongly related to T_C. These findings should give a valuable insight into the inhomogeneous magnetic properties of many DMS's [4].

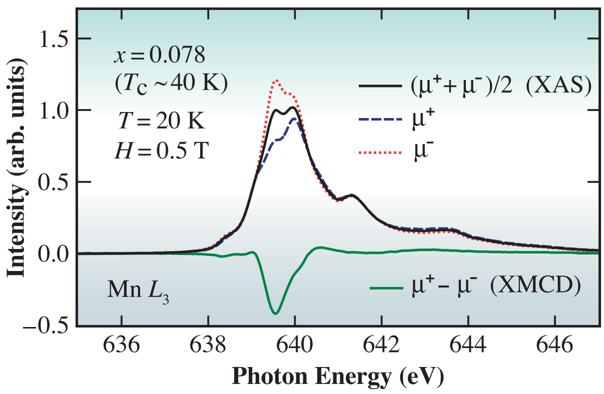

Fig. 2. XAS and XMCD spectra of x=0.078 at T=20 K and H=0.5 T in the photon energy region of the Mn L_3 absorption edge.

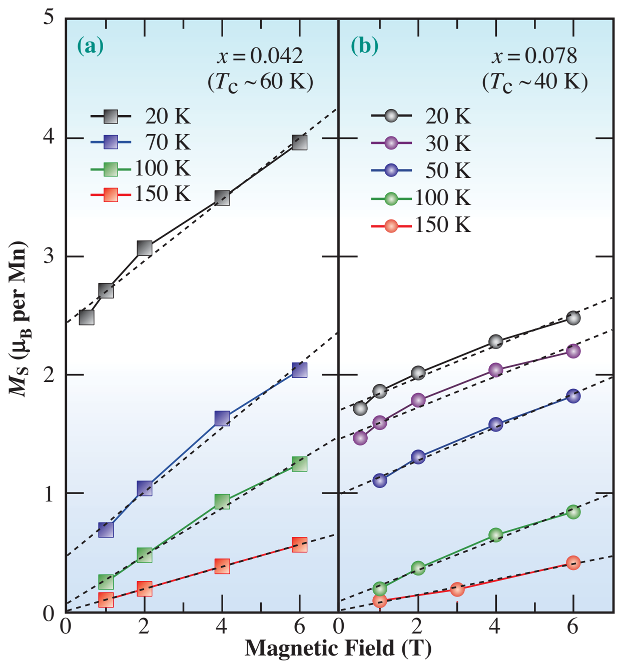

Fig. 3. H and T dependences of M_S **(a)** x=0.042 and **(b)** x=0.078. The dashed lines show fitted straight lines above H = 0.5 T.

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