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Perpendicular magnetic anisotropy of the ferromagnetic semiconductor $(Ba,K)(Zn,Mn)_2As_2$ studied by angle-dependent x-ray magnetic circular dichroism

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1 Introduction

Ferromagnetic semiconductors (FMSs) have attracted much attention in the field of spintronics since the discovery of ferromagnetism in Mn-doped III-V semiconductors such as (Ga,Mn)As and (In,Mn)As [1]. In such systems, ferromagnetic interaction is mediated by holes and, hence, can be controlled externally by changing the number of carriers through applying gate voltage or impinging light.

Recently, a new FMS $Ba_{1-x}K_x(Zn_{1-y}Mn_y)_2As_2$ was synthesized in bulk form [2], which crystalizes in the tetragonal $ThCr_2Si_2$ structure (I4/mmm) and is isostructural to 122-type Fe-based superconductors, as shown in Fig. 1. The host compound $BaZn_2As_2$ is a semiconductor with a narrow band gap of 0.2 eV. When holes and spins are introduced by K and Mn substitution for Ba and Zn, respectively, a ferromagnetic ground state is realized. With 30% of K and 15% of Mn substitution, the Curie temperature (T_C) reaches 230 K for polycrystalline samples (60 K for single crystals) and the carrier concentration 8×10^{20} cm^{-3}.

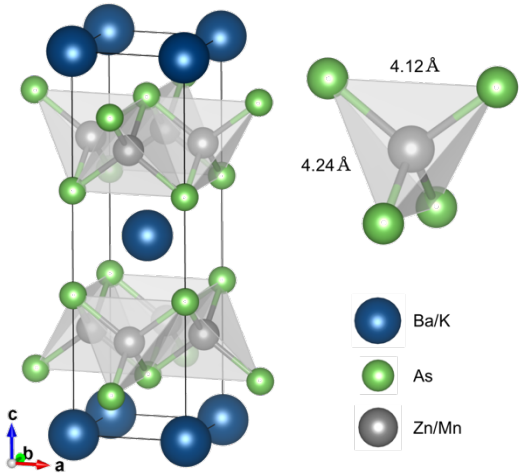

Fig. 1 Crystal structure of $(Ba,K)(Zn,Mn)_2As_2$.

Because the crystal structure is inherently anisotropic, where the Ba ions are located between the quasi-two-dimensional MnAs layers, and the $MnAs_4$ tetrahedrons are elongated to the c-axis by ~ 6% (see Fig. 1), sizable magnetic anisotropy would be expected. In fact, large perpendicular magnetic anisotropy was observed by SQUID measurements, which is advantageous for future applications such as magnetic memories. In the present study, we have investigated the magnetic anisotropy of this material by means of angle-dependent x-ray magnetic circular dichroism (AD-XMCD) measurements [3].

2 Experiment

$Ba_{0.904}K_{0.096}(Zn_{0.805}Mn_{0.195})_2As_2$ single crystals were grown by the arc-melting solid-state reaction method. AD-XMCD measurements were performed at BL-16A2 of Photon Factory, where we have developed a new apparatus equipped with two pairs of superconducting magnets so that magnetic fields up to 1 T can be applied to any direction. Prior to the measurements, the samples were cleaved *in situ* to obtain clean surfaces. Absorption signals were collected in the total-electron-yield mode and temperature was set to 20 K. Measurement geometry is shown at the top right of Fig.2.

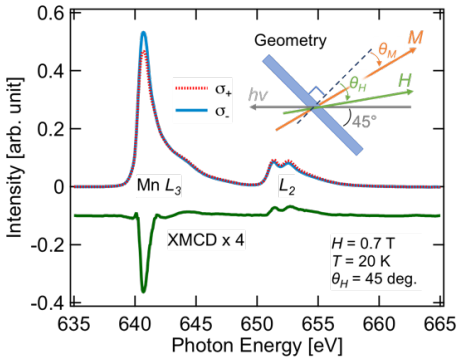

Fig. 2 Mn $L_{2,3}$-edge XAS and XMCD spectra of $Ba_{0.904}K_{0.096}(Zn_{0.805}Mn_{0.195})_2As_2$. Measurement geometry is also drawn at the top right.

3 Results and Discussion

Figure 2 shows XAS and XMCD spectra of $Ba_{0.904}K_{0.096}(Zn_{0.805}Mn_{0.195})_2As_2$ recorded at the Mn $L_{2,3}$ absorption edges. The XAS and XMCD spectra look similar to those of (Ga,Mn)As [4] and exhibit multiplet features. This suggests the localized nature of the Mn 3*d* electrons supporting the idea of carrier-induced ferromagnetism, where itinerant holes mediate ferromagnetic interaction between the localized spins. However, the magnetic moments of Mn ($m_{Mn} = m_{spin} + m_{orb}$) deduced using XMCD sum rules are about 0.4 μ_B at H = 0.7 T and T = 20 K, which is by far smaller than those of (Ga,Mn)As of 4.5 μ_B [4]. This implies the existence of antiferromagnetically coupled Mn pairs or magnetically inactive Mn atoms.

Figure 3 shows the deduced magnetic moments of Mn, projected onto light direction, as a function of magnetic field direction. If there is no magnetic anisotropy, where magnetic moments always point to the magnetic field direction, the data are expected to follow a sine curve as shown by the blue curve in Fig. 3. However, the data clearly deviate from the sine curve suggesting a considerable magnetic anisotropy in this system. Here, we have reproduced the data using Stoner-Wohlfarth model. In this model, the total energy of the system is expressed as [3]

$$E = \mu_0 M_{sat} H \cos(\theta_M - \theta_H) + (\mu_0 M_{sat}^2 - K_U)\cos^2\theta_M,$$

where μ_0 denotes the permeability of free space, M_{sat} the saturation magnetization, H the magnitude of the magnetic field and K_U the uniaxial magneto-crystalline anisotropy energy. As drawn in Fig. 2, θ_M and θ_H represent the angles of the magnetic moment and the magnetic field relative to the sample normal (c-axis direction). The first term represents the Zeeman energy, the second term the shape anisotropy energy and the third term the uniaxial anisotropy energy. From this equation, one can calculate θ_M (or the projected moment $M_{sat}\cos(45°\text{-}\theta_M)$) for given θ_H, H, M_{sat} and K_U by minimizing the total energy E. In this way, we have fitted the data treating K and M_{sat} as free parameters, and results are shown by the red curve in Fig. 3. The fit has yielded $K_U = 5 \times 10^5$ erg/cc and the saturation magnetization per Mn atom m_{Mn} = 0.40 μ_B. The positive value of K_U means that easy axis is along the c-axis.

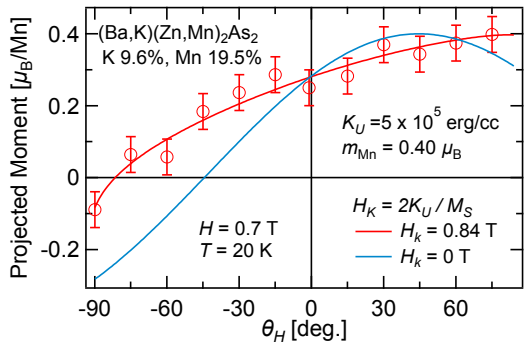

Fig. 3 Magnetic moments of Mn projected onto the light-incident direction as a function of magnetic field angle θ_H.

References

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