BL-16A/2023G046

Observing the orbital angular momentum of Fe and Co in chiral magnet $Fe_{1-x}Co_xSi$ (x = 0.25, 0.50) using soft x-ray magnetic circular dichroism

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The intermetallic compound $Fe_{1-x}Co_xSi$ has a helical magnetic order for 0.05 < x < 0.8, whereas its orbital angular momentum contributing to the occurrence of a Dzyaloshinskii–Moriya (D-M) interaction has not yet been verified. According to the previous neutron experiments, the magnitude of D-M interaction for x = 0.50 is approximately 60% of that for x = 0.25. We applied soft x-ray magnetic circular dichroism spectroscopy on the Fe $L_{2,3}$ and Co $L_{2,3}$ edges below critical temperature *T*c for x = 0.25 and 0.50 such that their orbital magnetic moments m_{orb} were evaluated independently. The ratio of m_{orb} to spin magnetic moment m_{spin} (m_{orb}/ m_{spin}) is independently estimated as (**1**) 3% for Fe and 9% for Co in x = 0.25 and (**2**) 3% for Fe and 11% for Co in x = 0.50. The m_{orb}/ m_{spin} ratio does not depend on the x value. The preset results suggest that the D-M interaction in $Fe_{1-x}Co_xSi$ indeed originates from the orbital angular momentum of Fe.

1 Introduction

The Dzyaloshinskii–Moriya (DM) interaction [1, 2] is driven by the spin–orbit coupling (SOC) and crystalline chirality. In general, SOC is weaker compared to other interactions (e.g., the crystalline field, orbital hybridization, and exchange interaction). The DM interaction arises from a combined second-order perturbation of SOC followed by an exchange interaction such that the DM is linear in the SOC. The DM interaction stabilizes a magnetic ground state with a spiral order of long wavelength. However, even within representative helical magnets, a quantitative understanding regarding the origin of the DM interaction, *i.e.*, the orbital angular momentum *L*, has not been sufficiently investigated except for a typical monoaxial chiral magnet, $CrNb_3S_6$ (space group $P6_322$) [3]. In $CrNb_3S_6$, the ratio of the orbital magnetic moment m_{orb} to the expected value of the spin magnetic moment m_{spin} is approximately 1%. As the second non-centrosymmetric target, we focus on a so-called B20 type material, $Fe_{1-x}Co_xSi$, which has two magnetic ions [4]. Both parent compounds FeSi and CoSi, as well as their mixtures, $Fe_{1-x}Co_xSi$, commonly have a B20 type crystal structure (space group $P2_13$), where all transition-metal sites are crystallographically equivalent. Indeed both parent compounds FeSi and CoSi are nonmagnetic. The insertion of Co into the Fe sites triggers the magnetic phase. For 0. 05 < x < 0.8, it is helical magnetic ordered, and the critical temperature *T*c is maximum at x = 0.35 [5–11], as seen in Fig. 1. Indeed the valences of Fe and Co in an intermetallic compound $Fe_{1-x}Co_xSi$ have yet to be determined. Both elements have the orbital magnetic moment m_{orb} and spin magnetic moment m_{spin} and participate in the formation of the helimagnetic order. Furthermore, a skyrmion lattice appears in the vicinity of *T*c [4, 11]. Thus, in terms of the development of ferromagnetic moments and DM interactions in a distorted alloy, $Fe_{1-x}Co_xSi$ is an interesting compound.

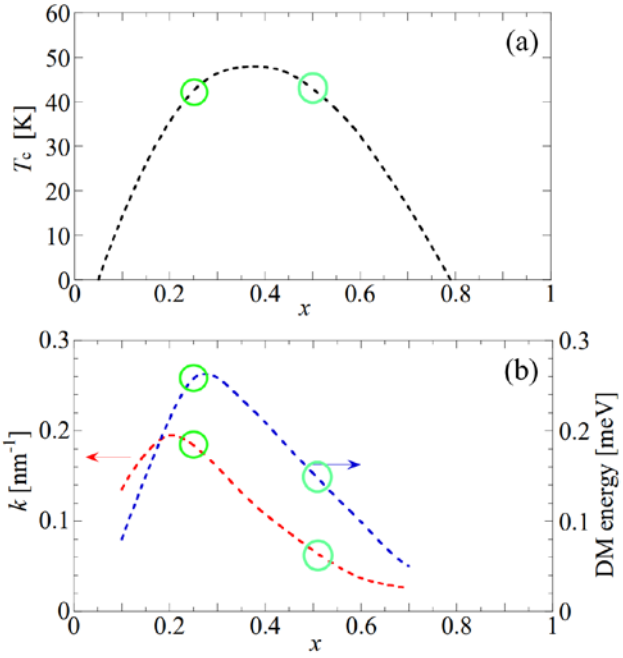

Fig. 1: x dependence of *T*c (a) [6–9, 11] and helix wave vector *k* and DM interaction (b) for $Fe_{1-x}Co_xSi$ [6–9, 11, 12]. The values corresponding to x = 0.25 and 0.50 are marked with open green circles.

2 Experiment

The XMCD spectroscopy is a powerful tool to obtain the magnetic information for selected elements, and furthermore the analysis of the XMCD spectrum yields the information of m_{orb} and m_{spin} for them. Soft XMCD spectroscopy was conducted on the beam line BL16A of KEK-PF. The photoabsorption spectra were obtained by directly measuring the intensity of the electron yield XMCD. The measurements were conducted below *T*c. Here, the magnetic field *H* was applied parallel to the [001] direction, parallel to the propagation vector of the helimagnetic modulation. The x-ray beam was nearly parallel to *H* of 1.2 T, which is much larger than the

critical magnetic field for the forced ferromagnetic state. The m_{orb} and m_{spin} values for Fe and Co were evaluated using the XMCD for the forced ferromagnetic state. The obtained results should be the same as those for other crystal orientations such as [110] and [111]. When H^+ and H^- are defined as parallel and antiparallel to the light, the deviation between the left (L)-handed μ_+ and right (R)-handed μ_- is observed under four settings: L-R at H^+, L-R at H^-, R-L at H^+, and R-L at H^-. The above series of energy scans was repeated several times and the data were averaged.

3 Results and Discussion

Figure 2 shows the XMCD spectra at H = 1.2 T for Fe (a) and Co (b) in $Fe_{0.75}Co_{0.25}Si$ [12]. As for Fe, the subtraction of the oxide contribution has been conducted. The integration of XMCD with respect to an increase in energy is also shown. After obtaining the integrated value of the XMCD value for L_3, L_2, and the whole L_3+L_2, we can estimate the m_{orb}/ m_{spin} ratio through the sum rule [13].

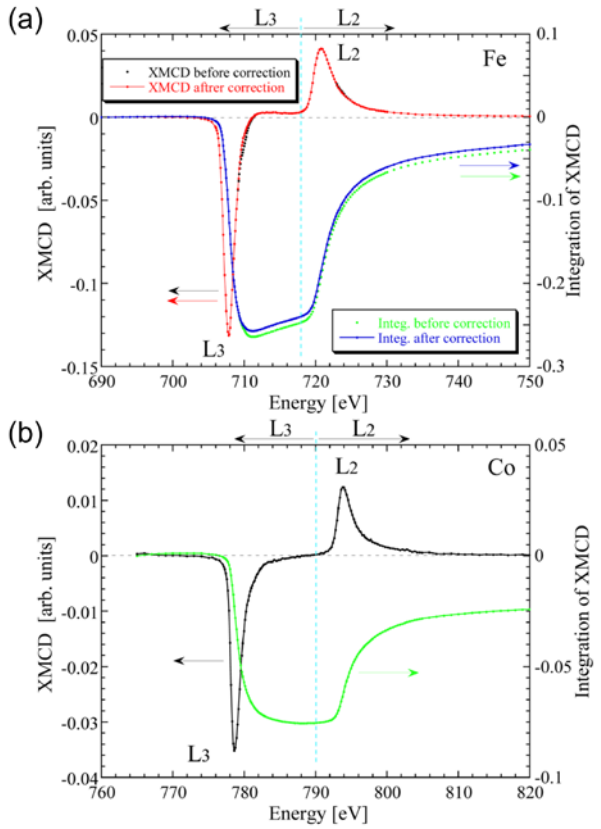

Fig. 2: XMCD spectra at H = 1.2 T for Fe (a) and Co (b) in $Fe_{0.75}Co_{0.25}Si$ [12]. The integration of XMCD with respect to an increase in energy is shown on the right vertical axis.

According to the previous neutron experiments, the magnitude of D-M interaction for x = 0.50 is approximately 60% of that for x = 0.25 as seen in Fig. 1. We conducted the soft x-ray MCD spectroscopy on the Fe $L_{2,3}$ and Co $L_{2,3}$ edges below Tc for x = 0.25 and 0.50. The ratio of m_{orb} to m_{spin} (m_{orb}/ m_{spin}) is independently estimated as (**1**) 3% for Fe and 9% for Co in x = 0.25 and (**2**) 3% for Fe and 11% for Co in x = 0.50. The m_{orb}/m_{spin} ratio does not depend on the x value. However, it is noted that the D-M interaction depends on the x value. The preset results suggest that the D-M interaction in $Fe_{1-x}Co_xSi$ originates from the orbital angular momentum of Fe.

Table 1: Analytic results on m_{orb}/ m_{spin} for $Fe_{1-x}Co_xSi$ with x = 0.25 and 0.50.

		m_{orb}/m_{spin} [%]
$Fe_{0.75}Co_{0.25}Si$	Fe	3.3
	Co	9.3
$Fe_{0.50}Co_{0.50}Si$	Fe	2.9
	Co	11.3

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