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Element-resolved magnetic moments of Heusler-type ferromagnetic ternary alloy Co_2MnGe

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Abstract

We have observed the site-resolved (Co, Mn and Ge) magnetic moments of Co_2MnGe with the use of 2p core absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD). The evaluated Co and Mn 3d spin magnetic moments are consistent with the values given by the neutron scattering experiment. We have found that the orbital magnetic moment is large for the Co 3d states and is recognizable even for the Mn 3d state, suggesting that a spin–orbit coupling should be reconsidered in the energy band structure in order to verify a half-metallic nature of this alloy. Moreover, we have revealed that the magnetic moment is induced on the Ge site in Co_2MnGe.

Recently, technologies to control the degrees of spin freedom have been considered for the next generation devices. The half-metallic ferromagnet, in which the majority spin density of states (DOS) crosses the Fermi level (E_F) while the minority spin DOS shows a semiconducting gap at E_F, is considered as one of the most useful candidates for tunnelling magneto-resistance (TMR) devices, because the magneto-resistance (MR) is expected to be divergently large if the conduction electron spin is 100% polarized [1, 2]. It is further expected that if we realize the material epitaxially grown on a semiconductor surface, 100% spin polarized electrons can be possibly injected into the semiconductor. Heusler-type Co_2MnGe with $L2_1$ structure is predicted to show the half-metallic energy band structure by Ishida *et al* for the bulk and the

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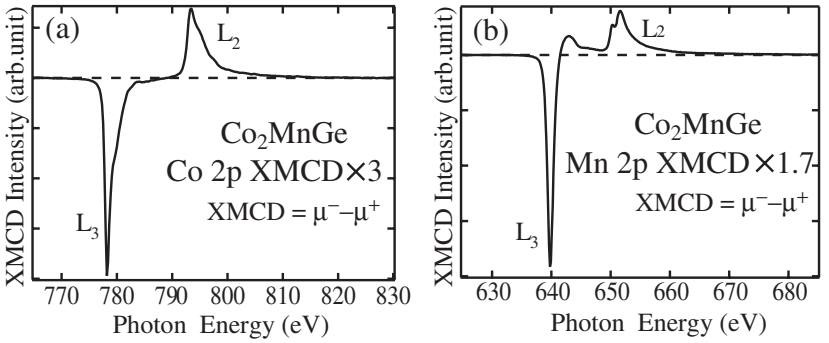

Figure 1. (a) Co and (b) Mn 2p XMCD spectra of Co_2MnGe.

thin film [3, 4]. This material is known to show a very large Curie temperature ($T_C \sim 905$ K) and huge magnetization ($M \sim 5\ \mu_B$/fu) [5]. Recently, Ambrose *et al* has reported that Co_2MnGe film can be grown on a GaAs substrate by a molecular beam epitaxy method [6]. Although several Heusler-type alloys are predicted to show the half-metallic energy band structures, the experimental reports on the evidence are little. This might be due to the existence of defects and the reduced symmetry at the surface or interface of the crystal [7]. On the theoretical side, the spin–orbit interaction is usually underestimated in the local density approximation (LDA), which might lead to modified electronic states. The calculated DOS of Co_2MnGe shows a rather small energy gap (∼0.5 eV) and the valence band maximum is located very close to E_F in the minority spin density of states [8]. Therefore, we suspect that the possible energy shift would derive much reduced electron spin polarization at E_F when the spin–orbit interaction is fully taken into account.

The Mn, Co and Ge 2p XAS and XMCD spectra for polycrystal of Co_2MnGe were measured on a helical undulator beamline BL25SU of SPring-8. The XAS spectra were measured by means of a total photoelectron yield. The measurement was performed with both the incident photon spin and the magnetization perpendicular to the sample surface. A external magnetic field of 1.4 T was applied. The XMCD spectra were taken by 1 Hz helicity switching of the incident circularly polarized light with fixed magnetic field [9]. The XMCD spectrum was defined as $\mu^- - \mu^+$, where μ^+ and μ^- represent the absorption intensity with the direction of magnetization being parallel and antiparallel to the photon spin (helicity), respectively. The measurement has been made under ultrahigh vacuum conditions ($<2.4 \times 10^{-8}$ Pa). A clean surface was obtained by *in situ* scraping of the sample surface with a diamond file. The temperature was about 45 K during the measurement, which was low enough compared to the Curie temperature ($T_C \sim 905$ K) [5]. The total magnetic moment of this sample was estimated to be 4.93 μ_B/fu from magnetization measurement at RT.

XMCD spectra in the Co and Mn 2p core excitation regions of Co_2MnGe are shown in figures 1(a) and (b). These line shapes are consistent with our former result [10], except for the larger XMCD magnitude in the present case. We have tried to evaluate the magnetic moments for the Co and Mn 3d electrons with the use of the sum rule [11, 12], where the term for the dipole operator is neglected since the averaged expectation value of the dipole operator can be close to zero for the polycrystalline sample [13]. The estimated spin magnetic moment (m_{spin}^{Co}) is 0.70 μ_B/Co atom assuming a Co 3d electron number of eight, which is given by the band structure calculation [14]. The estimation of the Mn 3d spin magnetic moment (m_{spin}^{Co}) from the sum rule is questionable because there is a substantial mixture of the $2p_{3/2}$ and $2p_{1/2}$ components due to their small energy separation [15]. Therefore, we have evaluated

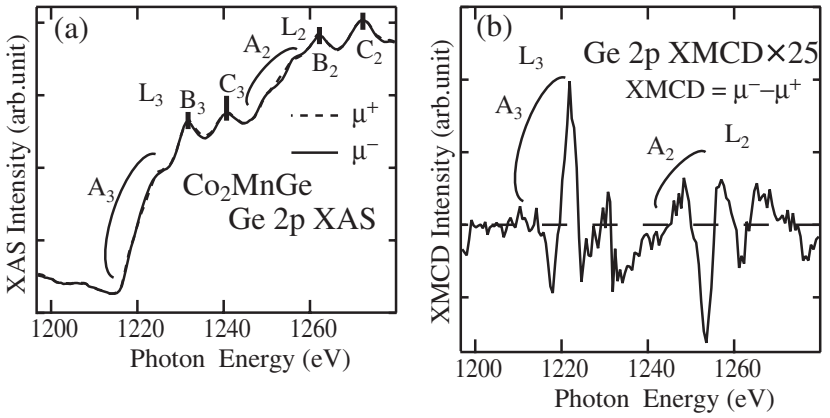

Figure 2. Ge 2p XAS (a) and XMCD (b) spectra of Co_2MnGe.

$m_{\mathrm{spin}}^{\mathrm{Mn}}$ of the Mn 3d state by subtracting $2\left(m_{\mathrm{spin}}^{\mathrm{Co}} + m_{\mathrm{orb}}^{\mathrm{Co}}\right)$ and $m_{\mathrm{orb}}^{\mathrm{Mn}}$ from the total magnetic moment ($\sim$4.93 μ_{B}/fu). Here, we assume that the magnetic moments of Ge 4sp electrons are negligibly small (but a finite moment is found as will be shown later). As a result, we obtain $m_{\mathrm{spin}}^{\mathrm{Mn}} = 3.40\ \mu_{\mathrm{B}}$/Mn atom. Moreover, we have considered that the total magnetic moment of Co_2MnGe at RT is equal to that at 45 K. These experimental values of m_{spin} for Mn and Co 3d states are consistent with the values observed by the neutron scattering [5]. It is noted that the experimentally obtained $m_{\mathrm{orb}}^{\mathrm{Co}}$ ($\sim$0.05 μ_{B}/Co atom) is about three times larger than that by the calculation ($\sim$0.02 μ_{B}/Co atom) [16]. The evaluated ratio $m_{\mathrm{orb}}/m_{\mathrm{spin}}$ of Co and Mn 3d states are 0.07 and 0.01. This shows that even the experimental $m_{\mathrm{orb}}^{\mathrm{Mn}}$ ($\sim$0.03 μ_{B}/Mn atom) is not negligible while the calculation shows only a tiny value. The probing depth of $\sim$2 nm for this measurement is large enough to detect bulk derived magnetic moments because their possible enhancement due to a broken translational symmetry could occur within a few monolayers ($<$1 nm) from the surface and the experiment usually shows the bulk derived value for thick films above 10 ML [17]. The present result on the substantial orbital magnetic moment suggests that the spin–orbit interaction of the Co 3d and even the Mn 3d electrons should be important for the electronic band structure of Co_2MnGe.

Next, the Ge 2p XAS and XMCD spectra are shown in figure 2. These spectra show rather complicated features. As one finds clearly, the energy region that contains the first three absorption structures (A_3, B_3, C_3) can be classified into the L_3 edge, while the second energy region including the next three features (A_2, B_2, C_2) corresponds to the L_2 edge. We find a clear XMCD signal that presents a negative–positive sign with increasing $h\nu$ in the region of A_3, while the signal with just the reversed sign is observed at the energy of A_2. We also find a small but finite XMCD in the higher energy regions of these features. It is clarified that the magnetic moment is induced on the Ge site possibly by the hybridization with the nearest neighbour Co atoms. We interpret that A_3 and A_2 are mainly caused by the Ge 2p–4s dipole transition. Here we show that these spectral features are understood in terms of the one-electron picture using one of the calculated results as follows [8]. By considering the transition probability, the XMCD spectrum would show a negative (positive) sign at the L_3 edge when the majority (minority) spin DOS is dominant. From the experimental result, we deduce that the majority spin DOS is dominant just above E_{F}, whereas the minority spin DOS exceeds the majority spin DOS in the higher energy region. In the energy region of A_3, the total magnitude of the XMCD signal at the L_3 edge shows the positive sign, suggesting a larger contribution of the

minority spin DOS compared to that of the majority spin in the unoccupied Ge 4s electronic states. This means that the Ge 4s spin magnetic moment couples parallel to those of the Mn and Co 3d electrons. Since the band structure in this higher energy region is not available, it seems to be difficult to discuss the other small XMCD features observed in the energy region including B_3 and C_3 for the L_3 edge and B_2 and C_2 for the L_2 edge. However, these features could possibly be caused by the Ge 2p–4d or 2p–5s intra-atomic excitations if one considers the weak interaction of Ge 4d5s states with the transition metal atoms.

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