Detected Quadrupole: A Hidden Source of Magnetic Anisotropy for Manganese Alloys

Mn-based alloys exhibit unique properties in spintronics materials that possess perpendicular magnetic anisotropy (PMA) beyond the Fe and Co-based alloys. Here, the origin of PMA in ferrimagnetic Mn_{3-x}Ga ordered alloys is investigated to resolve antiparallel-coupled Mn sites using X-ray magnetic circular and linear dichroism (XMCD/XMLD). We found that the contribution of orbital magnetic moments in PMA is small from XMCD and that the finite quadrupole-like orbital distortion through spin-flipped electron hopping is dominant from XMLD. These findings suggest that the spin-flipped quadrupole contributions originate from the PMA in Mn_{3-x}Ga and may lead to a paradigm shift in the research of PMA materials.

Perpendicular magnetic anisotropy (PMA) is desired for the development of high-density magnetic storage technologies. Recently, research using PMA films has focused on not only magnetic tunnel junctions toward spin-transfer switching magneto-resistive random-access memories but also antiferromagnetic or ferromagnetic devices. To design PMA materials, heavy-metal elements that possess large spin-orbit coupling are often utilized. However, the design of PMA materials without using heavy-metal elements is strongly desired and will be an important subject in future spintronics research. Mn-Ga binary alloys are a candidate that could overcome these issues. Mn_{3-x}Ga alloys with PMA satisfy the conditions of high spin polarization, low saturation magnetization, and low magnetic damping constants. Tetragonal Mn_{3-x}Ga alloys are widely recognized as having high PMA, ferromagnetic, or ferrimagnetic properties depending on the Mn composition. Two kinds of Mn sites, which couple antiferromagnetically, consist of Mn_{3-x}Ga. The L_21-type Mn_{3-x}Ga ordered alloy possesses a single Mn site (MnI). With increasing Mn concentration, ferrimagnetic coupling occurs by introducing an antiparallel MnII site as D_{022} symmetry. To investigate the mechanism of PMA and large coercive fields in Mn_{3-x}Ga, site-specific magnetic properties must be investigated explicitly. In this study, we performed X-ray magnetic circular and linear dichroism (XMCD/XMLD) measurements for Mn_{3-x}Ga to understand the PMA microscopically.

Samples with 3-nm-thick Mn_{3-x}Ga layers were prepared by magnetron sputtering on a 30-nm-thick Co_{2}Ga_{2} buffer layer using MgO (001) substrate [1]. The XMCD and XMLD were performed at BL-7A and BL-16A. For the XMCD measurements, a magnetic field of 1.2 T was applied, fixing photon helicities, parallel to the incident polarized beam. The total electron yield mode was adopted, and all measurements were performed at room temperature. In the XMLD measurements, the remnant states magnetized to PMA were adopted.

The Mn 2p-edge X-ray absorption spectra (XAS) and XMCD for Mn_{3-x}Ga with a single Mn site (MnI) are shown in Fig. 1. With increasing Mn concentration (decreasing x), the intensities of XAS increased and those of XMCD decreased because of the increase of antiparallel components. The element-specific magnetization curve at the Mn _L3-edge is also shown in the inset, showing PMA. The orbital magnetic moment values deduced from XMCD sum rules are too small to explain stabilization of the PMA because of the large magnetic crystalline anisotropy energy of the order of 10^5 J/m^2. Therefore, the magnetic dipole term (m_d) also stabilizes the PMA. To determine the effect of m_d, we performed XMLD measurements.

Figure 2 shows the linear polarization dependent XAS, where the directions are parallel and perpendicular to the sample magnetization as shown in the inset. The XMCD were detected by grazing incident XAS, where the directions are perpendicular and horizontal components. The element-specific magnetization curve at the Mn _L3-edge is also shown in the inset, showing PMA. The orbital magnetic moment values deduced from XMCD sum rules are too small to explain stabilization of the PMA because of the large magnetic crystalline anisotropy energy of the order of 10^5 J/m^2. Therefore, the magnetic dipole term (m_d) also stabilizes the PMA. To determine the effect of m_d, we performed XMLD measurements.

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