Detecting 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 $Mn_{3-\delta}Ga$ 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 orbital quadrupole formations originate from the PMA in $Mn_{3-\delta}Ga$ 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 ferrimagnetic 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. $Mn_{3-\delta}Ga$ alloys with PMA satisfy the conditions of high spin polarization, low saturation magnetization, and low magnetic damping constants. Tetragonal $Mn_{3-\delta}Ga$ 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 $Mn_{3-\delta}Ga$. The $L1_0$-type Mn_1Ga ordered alloy possesses a single Mn site (MnI). With increasing Mn concentration, ferrimagnetic coupling occurs by introducing an antiparallel MnII site as $D0_{22}$ symmetry. To investigate the mechanism of PMA and large coercive fields in $Mn_{3-\delta}Ga$, site-specific magnetic properties must be investigated explicitly. In this study, we performed X-ray magnetic circular and linear dichroism (XMCD/XMLD) measurements for $Mn_{3-\delta}$ Ga to understand the PMA microscopically.

Samples with 3-nm-thick $Mn_{3-\delta}Ga$ layers were prepared by magnetron sputtering on a 30-nm-thick $Co_{55}Ga_{45}$ buffer layer using MgO (001) substrate [1]. The XMCD and XMLD were performed at BL-7A and 16A. For the XMCD measurements, a magnetic field of ±1.2 T was applied, by 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.

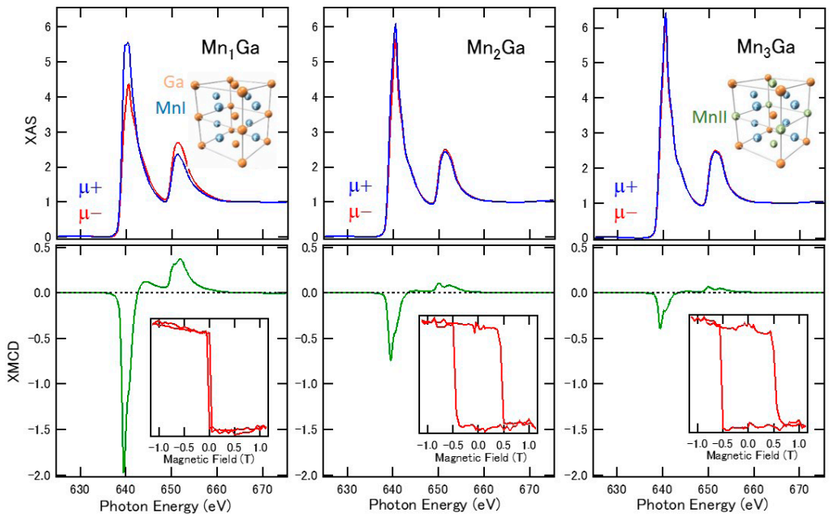

Figure 1: XAS and XMCD of $Mn_{3-\delta}Ga$ for δ = 0, 1, and 2. μ+ and μ− denote the absorption in different magnetic field directions. The insets show the crystal structure of $L1_0$ and $D0_{22}$ symmetries and the magnetic field dependence of the hysteresis curves taken by fixed L_3-edge photon energy.

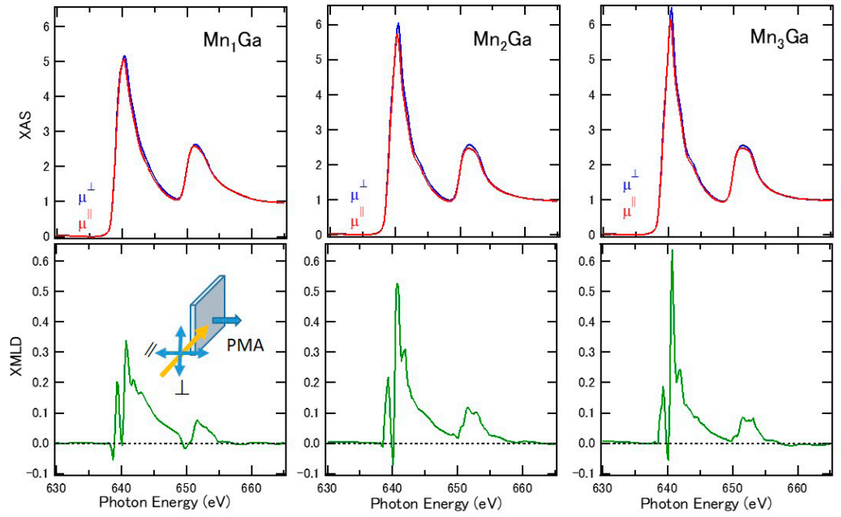

Figure 2: XAS and XMLD of $Mn_{3-\delta}Ga$ for δ = 0, 1, and 2. $\mu^{\perp}$ and $\mu^{\parallel}$ denote the absorption in different electric field directions. The inset shows an illustration of the XMLD measurement geometry.

The Mn *L*-edge X-ray absorption spectra (XAS) and XMCD for Mn_1Ga with a single Mn site (MnI) are shown in Fig. 1. With increasing Mn concentration (decreasing δ), 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 L_3-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 10^6 J/m^3. Therefore, the magnetic dipole term (m_{Tz}) also stabilizes the PMA. To determine the effect of m_{Tz}, we performed XMLD measurements.

Figure 2 shows the linear polarization dependent XAS, where the directions are perpendicular and horizontal to the sample magnetization as shown in the inset. The XMLD were detected by grazing incident beams tilted 60° from the incident beam. With increasing Mn composition, the XMLD signal intensities were enhanced because XMLD detects the square of magnetization M^2 contribution. In Mn_3Ga, XMLD includes the summation of both $M^2_{MnI} + M^2_{MnII}$ contributions. We note that the integrals of the XMLD line shapes are proportional to quadrupole Q_{zz} along the direction normal to the sample surface. We confirmed that the integral converges to a positive value, deducing that the sign of Q_{zz} is positive with the order of 0.01 by applying the XMLD sum rule in the notation of $m_{Tz} = -Q_{zz}\cdot S$ with spin angular momentum *S*; that is, magnetized $3z^2$-r^2 orbitals are strongly coupled with the incident beam and are elongated in an easy-axis direction. These suggest the orbital polarization of Mn 3*d* states along the *z*-axis direction results in the formation of the cigar-type prolate unoccupied orbital orientation, which causes the PMA in $Mn_{3-\delta}Ga$ [2]. These results are also reproduced from density-functional-theory calculations. Therefore, combining both XMCD and XMLD, we found that the contribution of m_{Tz} is essential for stabilizing the PMA, which suggests that the quadrupole-like spin-flipped states through the anisotropic $L1_0$ and $D0_{22}$ crystalline symmetries originate in the PMA in $Mn_{3-\delta}Ga$. The present study provides a promising strategy to investigate quadrupoles in antiferro or ferrimagnetic materials with PMA.

REFERENCES

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[2] J. Okabayashi, Y. Miura, Y. Kota, K. Z. Suzuki, A. Sakuma and S. Mizukami, *Sci. Rep.* **10**, 9744 (2020).

BEAMLINES

BL-7A and BL-16A

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