Perpendicular orbital and quadrupole anisotropies at Fe/MgO interfaces detected by x-ray magnetic circular and linear dichroisms

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MgO-based magnetic tunnel junctions (MTJs) have been exploited toward an ultrahigh tunnel magnetoresistance (TMR) through the large interfacial PMA between MgO and transition-metal (TM) alloys. The PMA of 1.4 MJ/m^3 at the interfaces between MgO and Fe has been achieved [1]. As an understanding of PMA induced at the interface between the ferromagnetic layer and the MgO barrier layer, the electronic and magnetic structures of the interfaces have to be clarified explicitly. In order to evaluate the PMA energy, it is necessary to elucidate the anisotropy of orbital magnetic moments along parallel and perpendicular directions to the surface by x-ray magnetic circular dichroism (XMCD) [2]. A recent study of the theoretical calculations suggests the existence of the quadrupole tensor (Q_{ij}) at the Fe/MgO interface through the interfacial strain due to the lattice mismatching of 3.8% between Fe and MgO [3]. Therefore, the understanding of interfacial orbital magnetic moments and quadrupole moments at Fe/MgO is necessary for unveiling the Fe/MgO-based PMA and further functional interfacial PMA designing. Here, we report the linear dichroism (XMLD) technique adopting for PMA at single crystal Fe/MgO(001) interfaces and discuss the effect of Q_{ij} for the PMA beyond the anisotropic orbital moments.

Samples were grown by using ultrahigh vacuum electron-beam evaporation on MgO(001) substrates. The XMCD and XMCD were performed at BL-7A and 16A, respectively, in the Photon Factory (KEK). The total electron yield mode was adopted, and all measurements were performed at room temperature. For the XMLD measurements, the sample was magnetized along the sample surface normal easy-axis direction and reversed to zero to use the remanent states and was rotated to the grazing incidence geometry.

Figure 1 shows the x-ray absorption spectra (XAS) of a 0.7-nm-thick Fe/MgO interface by different circular [Fig. 1(a)] and linear [Fig. 1(b)] polarized beams. The XAS line shapes show clear metallic states without oxidation are detected. Using the XMCD sum rules, m_{orb} and m_s^{eff} are estimated to be 0.21 and 1.79 μ_B, respectively, for the normal component. Although m_s is smaller than that in bulk, m_{orb} is enhanced at the interface. The inset of Fig. 1 shows the *M-H* curve in Fe L_3-edge XMCD for the easy-axis direction. Clear square-shape hysteresis coincides with the magnetization curves measured by SQUID.

The XMLD sum rule suggests that the integrals of XMLD line shapes are proportional to Q_{zz}. We confirmed in Fig. 1(b) that the integral of XMLD intensities converges to positive, which is related to the sign of Q_{zz} in dichroism to be positive; that is, unoccupied $3d_{z^2}$ orbitals are slightly elongated to the out-of-plane direction. We note that XMLD is defined as ($\boldsymbol{M} \perp \boldsymbol{E}$) – ($\boldsymbol{M} /\!/ \boldsymbol{E}$), where $\boldsymbol{M}$ and $\boldsymbol{E}$ is the magnetization and the electric filed vector of incident synchrotron, respectively, resulting that the positive XMLD means preferential coupling with the out-of-plane unoccupied orbitals. However, since the denominator in the sum rule is two orders of magnitude larger than that of the numerator, the absolute value of Q_{zz} is estimated to be less than 0.01, which is interpreted as less than 1% unoccupied orbital polarization of Fe 3*d* states. On the other hand, electron occupancies are slightly shrinking through the relation $Q_{xx}+Q_{yy}+Q_{zz}=0$. The XMCD with the angular dependence also deduces the quite small magnetic dipole term m_{T} using the spin sum rule. These results are consistent with the fact that the contribution of m_{T} is two orders of magnitude smaller than that of m_s (1.73 μ_B) [4]. We note that the notation of Q_{zz} is independent of spin-orbit coupling of Fe, suggesting that the origin of Q_{zz} is derived from the interfacial asymmetry and strain.

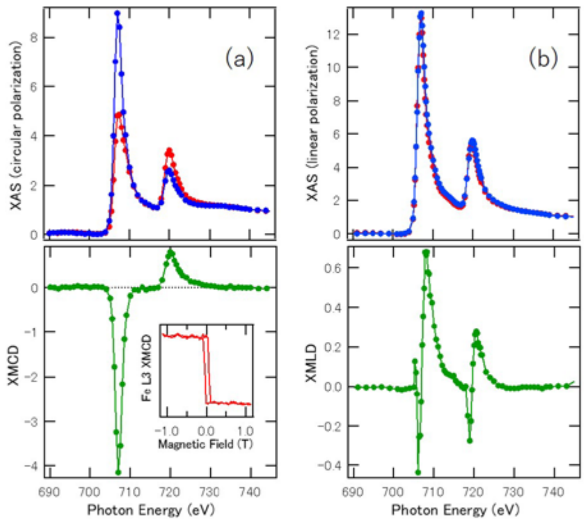

FIG. 1. (a) XAS and XMCD by circularly polarized beams
(b) XAS and XMLD by linearly polarized beams for Fe L-edges at the Fe /MgO interface. The inset is the XMCD hysteresis curve at the Fe L_3-edge for surface normal direction [4].

References

[1] J.W. Koo et al., *Appl. Phys. Lett.* **103**, 192401 (2013).
[2] J. Okabayashi et al., *Appl. Phys. Lett.* **105**, 122408 (2014).
[3] K. Masuda and Y. Miura, *Phys. Rev. B* **98**, 224421 (2018).
[4] J. Okabayashi et al., *Appl. Phys. Lett.* **115**, 252402 (2019);

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