

Microscopic origin of magnetostriction in Fe₃Ga studied by *operando* XMCD

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The large magnetostriction (MS) effect has garnered attention for device applications involving sensors or vibrational power generation [1]. Although most significant MS effects depend on materials containing rare-earth elements, Fe₃Ga (Galfenol) modifies Fe-based materials to achieve a large MS of 300 ppm, which is ten times greater than that of conventional alloy compounds. However, the reason for the large MS in Fe₃Ga remains unclear. The doping of Ga atoms into pure bcc-Fe enhances the MS by altering the electronic structures [2]. The piezoelectric control of Fe₃Ga/ferroelectric materials has also gained interest through the introduction of reversible strain. From the perspective of electronic structures, the spin and orbital states under applied reversible strain must be examined. We have developed electric field (*E*)-induced *operando* x-ray magnetic circular dichroism (XMCD) technique to apply *E* to the ferroelectric substrate Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ (PMN-PT), which reversibly tunes the interfacial lattice constants of the Fe₃Ga magnetic layer [3]. In this study, we discuss the microscopic origin of inverse magnetostriction effects or orbital-elastic effects in Fe₃Ga concerning the orbital magnetic moments (*m*_{orb}) using *E*-induced XMCD. Furthermore, to investigate the strain propagation into the Fe₃Ga layer induced by the piezoelectric effect in PMN-PT, extended X-ray absorption fine structure (EXAFS) analysis at the Fe *K*-edge is utilized to comprehend the element-specific modulation of the lattice distances.

We prepared samples of a 10-nm-thick Fe₃Ga (422) layer grown on single-crystal PMN-PT(011) substrates, incorporating a 0.3-nm-thick Fe layer via molecular beam epitaxy. The *E*-induced modulation of the in-plane magnetic properties was characterized using X-ray absorption spectroscopy (XAS) and XMCD, while the structural modulation due to *E* was monitored through EXAFS. *Operando* XMCD and EXAFS measurements were conducted at BL-7A and 9A, respectively, in the Photon Factory (KEK). The partial-fluorescence-yield mode was employed for both experiments. To apply an *E* to the PMN-PT substrate along the [011] direction, a Au(100 nm)/Ti(3 nm) electrode was deposited on the backside of the PMN-PT substrate, with the Fe₃Ga film serving as the top electrode. For XMCD measurements, the photon helicity was fixed while the magnetic field and beam incidence direction were varied for both positive and negative values.

The magnetization measurements indicated that the magnetic easy axis is slightly altered when applying *E* due to the modulation of in-plane lattice strain of approximately 0.1% in PMN-PT. Figure 1 displays the XAS and XMCD under applying *E*. XAS line shape exhibits clear metallic

shapes and XMCD at *L*₃ slightly modulated by applying *E*. This originates from the changes in *m*_{orb}. By applying an *E* of −8 MV/m, the magnetic-field dependence at *L*₃-edge shown in Fig. 1(c) clearly shows the changes of magnetic easy and hard axes direction. The saturation magnetization value increased in the magnetic-field dependence of Fe *L*-edge in XMCD spectral analysis. The variations in *L*₃-edge XMCD intensity arise from the in-plane lattice strain. The increase in *m*_{orb} due to *E* mirrors the phenomenon seen in Co₂FeSi/PMN-PT [4] and differs from that in Fe₃Si/PMN-PT. Given that the electronic structures in Fe₃Ga derived from band-structure calculations differ from those in Fe, *m*_{orb} in Fe₃Ga is influenced by strain, resulting in a significant MS effect in Fe₃Ga. Additionally, Fe *K*-edge *operando* EXAFS also detected the modulation of nearest neighbor distances in Fe sites. Mössbauer spectroscopy further confirms the modulation of *m*_{orb}. Therefore, applying reversible strain through *E* to Fe₃Ga allows for the monitoring of orbital striction, clarifying the microscopic origin of the significant MS effects in Fe₃Ga, which can only be detected by *operando* XMCD and EXAFS.

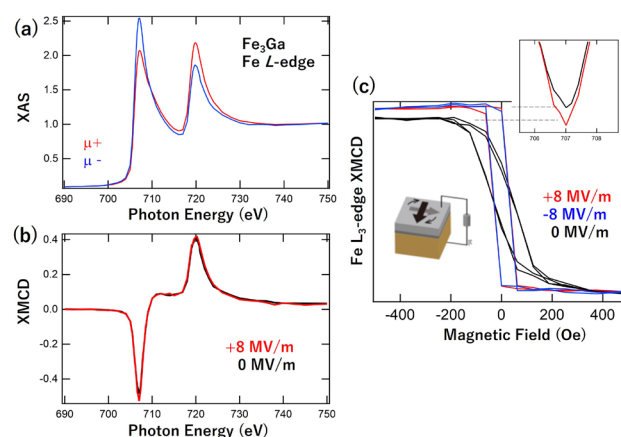


Fig. 1, XAS (a) and XMCD (b) under an applied electric field in Fe₃Ga/PMN-PT. (c) Magnetic-field dependence of Fe *L*₃-edge XMCD under an applied *E* of ± 8 MV/m with reversibly switching the applying electric field.

References

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