Orbital Elasticity in an Interfacial Multiferroics of Co_2FeSi on Ferroelectric Substrates Studied by Operando X-Ray Spectroscopies

Artificial ferromagnetic/ferroelectric multiferroic hetero-structures have been widely investigated for the control of magnetism and magnetic anisotropy by applying electric field, which is essential for the low-power operation device fabrications. To investigate the origin of electric field control, we developed a method for X-ray absorption and magnetic circular dichroism spectroscopies by applying piezo electric fields to the ferroelectric material. For the interface between Heusler alloy Co_2FeSi and ferroelectric material, we report the modulation of orbital magnetic moments by reversible tensile and compressive strains through the electric field, which results in the novel phenomena of 'orbital elasticity'.

Combining ferromagnetic and ferroelectric properties has been investigated as multiferroics, which provides considerable advantages of magnetization control by an electric field (E) as a low-energy power consumption operation without electric current. As spintronics research pursues the suppression of power consumption for device operations, the manipulation of the spins by E is one of the solutions for future device applications. Recent developments have focused on the modulation of magnetization by a low electric field. Recently, a study reported that Co_2FeSi / $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) interfacial multiferroics exhibited a giant magneto-electric (ME) effect of 1.8 x 10^{-5} s/m, where Co_2FeSi (CFS) is a Co-ased Heusler alloy that was expected to have high spin polarization at room temperature and high Curie temperature [1]. The origin of the large ME effect in CFS-based interfacial multiferroics was associated with strain-mediated magnetic anisotropy modulation. However, the microscopic origin of the giant ME effects in the CFS/PMN-PT interfacial multiferroics is still unknown. To initiate novel research into the physics of the relationship between the lattice distortion and orbital magnetic moments (m_{orb}), the anisotropic m_{orb} needs to be explored even in interfacial multiferroics. Since *operando* X-ray magnetic circular dichroism (XMCD) can probe element-specific orbital modulation when a reversible strain

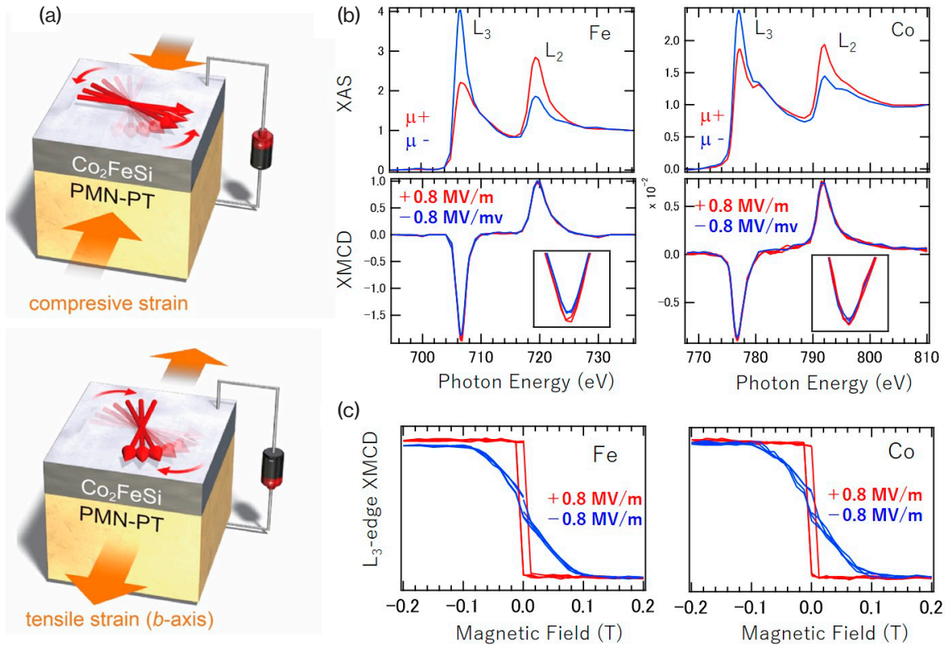

Figure 1: (a) Schematic illustrations of the changes in the magnetic anisotropy by an applied E in the strain directions. **(b)** XAS and XMCD under an applied electric field in Co_2FeSi/PMN-PT at Fe and Co L-edges at ±0.8 MV/m. Expanded views around L_3 edges are shown in the bottom panels. **(c)** Magnetic field dependence of Fe and Co L_3-edge XMCD under an applied electric field.

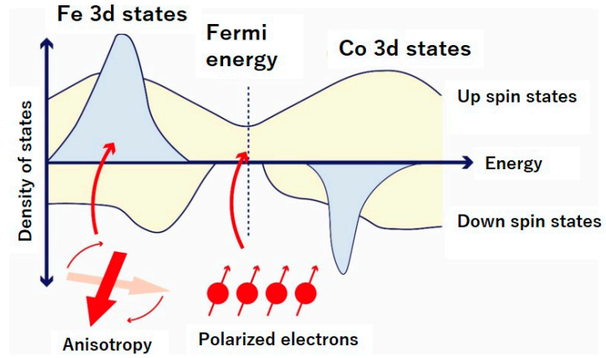

Figure 2: Schematic illustration of the density of states in Co_2FeSi.

is applied, we developed the XMCD system with applying E. To investigate the element-specific structural changes by applying strain, operando extended X-ray absorption fine structures (EXAFS) was also employed.

X-ray absorption spectroscopy (XAS) and XMCD measurements of the Fe and Co L-edges were performed using the BL-7A beamline, at room temperature [2, 3]. *Operando* EXAFS using Fe and Co K-edges were also performed at BL-9A. The electrodes were mounted on the surface of the sample as ground and E was applied from the backside of the substrate during the measurements.

Figure 1 shows the Fe and Co L-edge XAS and XMCD spectra obtained in the partial fluorescence yield mode under an applied electric field E of ±0.8 MV/m. The XAS spectral line shapes originated from metallic Fe and Co features in CFS. The spectral line shapes of XMCD and the slight change in the L_3-edge peak were modulated by E only at the Fe L-edge, despite the fixed sample measurement position. These results indicated that the m_{orb} were modulated by the applied E, thereby resulting in changes in magnetic anisotropy. The modulation of m_{orb} by 0.01 μ_B under an applied E was related to the induced lattice distortion from the PMN-PT substrates. In contrast to the case of Fe, no spectral changes were detected for the Co L-edge XMCD. Furthermore, *operando* EXAFS also detected the changes of nearest neighbor distances by applying E through the tensile (positive E) and compressive (negative E) strains for both Fe and Co sites.

The element-specific XMCD hysteresis curves at the Fe and Co L_3-edges during the application of E. In the case of +0.8 MV/m, easy axis behavior was clearly observed, whereas the −0.8 MV/m case indicated the hard axis. This result showed reproducible behavior of the changes in the magnetic easy-axis direction. In the case of Fe shown in **Fig. 1(c)**, the vertical axis strength was slightly different between ±0.8 MV/m, thus affecting the changes in XMCD in the Fe L_3-edge intensity, whereas the hysteresis curves of Co by ±0.8 MV/m overlapped above the saturation magnetic fields (**Fig. 1(c)**). Therefore, only the change in m_{orb} for Fe was experimentally found.

In Co-based Heusler alloy compounds, the density of states (DOS) of Co is generally dominant at the Fermi level for half-metallic conductivity, as shown in **Fig. 2**; however, the DOS of Fe is located deep in the valence band and thus contributes to the local magnetic moments, which is the reasons for the m_{orb} changes reliance only on the Fe sites. The nonmagnetic element Si can be valid for structural stability and tuning of the energy band gap and the position of the Fermi level. The role of each element is relevant in the CFS layer and can be tuned by strain. Therefore, the control of the m_{orb} only in Fe with an applied strain is plausible for the magnetic anisotropy control in Co-based Heusler alloy compounds. These element-specific orbital elasticities can be achieved only by *operando* XMCD and EXAFS [4].

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BEAMLINES

BL-7A and BL-9A

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