Magnetoelectric-effect-driven antiferromagnetic domain motion revealed by scanning soft X-ray magnetic circular dichroism microscopy

Magnetic materials are classified roughly into ferromagnets and antiferromagnets. Ferromagnets have spontaneous magnetization and have been a major player in both magnetic research and various magnetic devices. By contrast, in antiferromagnets, magnetic moments are fully compensated and no net magnetization emerges. Because of this inherent characteristic, antiferromagnets have long been regarded as useless materials. However, once the antiferromagnetic (AFM) spin, especially in micro/nanosize devices, can be controlled by some external field, we can enjoy various functionalities such as the ultrafast AFM spin/domain dynamics, which yield novel phenomena such as THz emission. Owing to the recent development of spintronics, spin-torque and magnetoelectric (ME) effects are expected to become a key to the control of the AFM spin/domain. The ME effect is referred to as magnetization (\mathbf{M}) induction by an electric field (E) and electric polarization (P) by a magnetic field (**H**) through the ME susceptibility $\alpha :: \mathbf{M} = \alpha' \cdot \mathbf{E}$ and $P = \alpha \cdot H$. Cr₂O₃ is a prototypical ME-AFM material known since the middle of the1950s. In 2005, Borisov et al. demonstrated that the ME-controlled AFM domain state in Cr₂O₃ was detectable through the exchange bias polarity, which was induced by the interfacial exchange coupling between the AFM Cr₂O₃ and ferromagnetic (FM) layers [1]. In this scenario, it is assumed that the Cr₂O₃ AFM domain, or at least the interfacial Cr spin, FM domain, couples with Cr₂O₃ and exchange bias polarity are connected mutually; this has been believed but is unproven. We prove that direct evidence to support this hypothesis can be obtained through magnetic domain observation using the scanning soft X-ray magnetic circular dichroism (XMCD) microscope equipped at SPring-8 BL25SU [2] by taking advantage of the elementspecific feature. In this instrument, the circularly polarized soft X-ray is focused on the sample using the Fresnel zone plate, and the sample is scanned two dimensionally to maintain the focused state. During scanning, the soft X-ray absorption intensity is collected. By subtracting the obtained image collected with the opposite helicity of the incident soft X-ray, two-dimensional mapping of the XMCD intensity can be accomplished.

Figure 1 shows the two-dimensional map of the XMCD intensity measured at H = 0 T with the incident photon energy of the (a) Co L_3 -edge (778.6 eV) and (b) Cr L_3 -edge (576.1 eV) for a Pt 1.5 nm/Co 0.4 nm/Au 1.0 nm/Cr₂O₃ 150 nm/Pt 20 nm thickness multilayer film [3]. Upward and downward Co FM domains with several μ m are visualized in Fig. 1(a). Similar contrast is observed in Fig. 1(b), indicating that a significant XMCD signal can

be obtained from the interfacial Cr spin. It is noteworthy that the two-dimensional distribution is spatially coupled with the Co FM domain. Additionally, the *H*-dependence of XMCD intensity measured on an individual domain (not shown) revealed that the sign of exchange bias polarity is opposite for oppositely directed domains. In the following (Figs. 2 and 3), on the basis of this finding, the (quasi-) static AFM domain wall motion is explored through the magnetic domain observation of FM Co, i.e., XMCD imaging at the Co L_3 -edge.

Other important but difficult issues are the switching process [3] and dynamics [4]. The switching process of magnetic materials is either coherent rotation or magnetic domain wall propagation. In a general FM substance, either switching mode can occur, depending on the size of the magnetic substance; below and above the characteristic size, which is on the order of the magnetic domain wall width, coherent rotation or magnetic domain wall propagation occurs, respectively. In the case of the AFM substance, it is not established whether a similar switching process occurs, although this is widely believed. We address this issue by directly observing the change in the magnetic domain pattern on applying a ME field. For such observation, there are many technical requirements, e.g., high magnetic field (> a few T), temperature controllability, and in situ E application, all of which are met by the aforementioned instrument.

Figure 2 shows the magnetic domain pattern in the remanent state, i.e., H = 0 T and E = 0 MV/m, after applying various *E* with constant H = +4 T. In the switching process, the newly nucleated magnetic domains are rarely observed but the expansion of the pre-existing magnetic domains are observed; the red (blue) magnetic



Fig. 1. Two-dimensional mapping of XMCD intensity measured at the photon energy of the Co L_3 -edge (top right) and Cr L_3 -edge (bottom right). Blue and red regions correspond to upward and downward magnetizations, respectively. The measurement temperature was 280 K and *H* during the observation was zero. In the left side of the figure, the stacking structure of the studied film is shown.



Fig. 2. Magnetic domain pattern after applying ME field, with H = +4 T and various *E*, showing increasing (top) and decreasing (bottom) branches of *E*. Observations were done at 280 K. The photon energy used for the imaging was 778.6 eV (Co L_3 -edge). In the top left of the figure, an optical microscope image of the fabricated Hall device with the equivalent circuit diagram used to apply *E* is shown. The Hall device is 10 µm wide and 50 µm long. In this device, the 20-nm-thick Pt buffer layer and the Pt/ Co/Au layer were used as the bottom and top electrodes, respectively, to apply *E* to the Cr₂O₃ layer. After applying *E* and *H*, both fields were removed during the XMCD measurements. A bias voltage, which is often applied to improve the signal-to-noise ratio in the TEY method, was not applied during the measurements. *E* and *H* were applied in the direction perpendicular to the film and the positive direction was defined as the direction from the substrate side to the film side.

domain expands in the increasing (decreasing) branch, which indicates that magnetic domain wall propagation is predominant. Referring to the finding in Fig. 2, we further address the ME-induced magnetic domain wall dynamics of the AFM Cr_2O_3 . In contrast to the current-induced magnetic domain wall propagation in the FM materials, the propagation direction of the AFM domain wall is difficult to define by *E*. The direct observation of the magnetic domain pattern is a way to directly address this issue.

Figure 3 shows the evolution of the magnetic domain pattern measured at zero ME field after applying a pulsed ME field. The applied ME field condition before the domain observation is similar to that in Fig. 2 except that pulsed *E* with an amplitude of -133 MV/m and various widths were applied instead of DC *E* in the case of Fig. 2. Details of the measurement procedure can be found in our previous report [4]. Starting from the initial state, the blue magnetic domain expands with increasing pulse width. The magnetic domain wall propagation as a function of the pulse width gives the AFM domain wall velocity *v* driven by the ME effect: $v \sim 22$ m/s in the case of Fig. 3. *v* depends on the amplitude of *E* and the v-Erelationship should yield a variety of ME-driven AFM spin/ domain dynamics such as the effective Gilbert damping parameter of the AFM Cr_2O_3 . To meet such growing interest, detection of the AFM order parameter without using exchange coupling with the FM moment is desirable because the low spin dynamics of the AFM spin.



Fig. 3. Evolution of magnetic domain after applying ME field with pulse E = -133 MV/m with various pulse widths and H = +4 T. Observations were done at 280 K. The photon energy used for the imaging was 778.6 eV (Co L_3 -edge). The definition of blue and red domains and the measurement sequence are similar to those of Fig. 2.

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