Voltage-Induced Changes in Magnetism of FeCo/BaTiO₃ Thin Films Studied by X-Ray Absorption Spectroscopy*

Kenta Amemiya[†] and Masako Sakamaki

*Photon Factory and Condensed Matter Research Center,
Institute of Materials Structure Science, High Energy Accelerator Research Organization,
Oho 1-1, Tsukuba, Ibaraki 305-0801, Japan, and*

*Department of Materials Structure Science, School of High Energy Accelerator Science,
The Graduate University for Advanced Studies (SOKENDAI), Oho 1-1, Tsukuba, Ibaraki 305-0801, Japan*
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Voltage-induced changes in the magnetic properties of FeCo thin films grown on a BaTiO₃ substrate are investigated by means of the X-ray magnetic circular dichroism (XMCD). The magnetization curves show different effects of the applied voltages depending on the Fe-Co compositions, which are ranging from Fe₆₇Co₃₃ to Fe₂₅Co₇₅. The XMCD spectra suggest that magnetization of the Fe₅₀Co₅₀ film at the saturation state are different between the positive and negative applied voltages, while those for other compositions have no detectable dependence on the applied voltage. The peculiar behavior for Fe₅₀Co₅₀ could be related to the abrupt change in the density of state at the Fermi level. [DOI: 10.1380/ejsnt.2015.465]

Keywords: Magnetic Films; Dielectric Substrate; Voltage Effect; X-ray Absorption Spectroscopy

I. INTRODUCTION

The control of the magnetic direction by voltage application is one of the most promising methods to reduce the energy consumption in the magnetization switching process. In fact, the voltage-induced magnetization switching was recently demonstrated for FeCo thin films [1]. The idea to control magnetization by an electric field has been proposed as a multiferroic concept [2], which is realized, for instance, in one of the most famous multiferroic materials, BiFeO₃. However, the temperature at which such materials show the multiferroic property is usually much below the room temperature (RT) in the case of single-compound materials such as BiFeO₃. On the other hand, it was reported that the coercive field of ferromagnetic Fe thin film grown on a ferroelectric BaTiO₃ (BTO) substrate changes when a voltage is applied between the Fe film and the bottom of the substrate even at RT [3]. Moreover, we have observed a slight change in the X-ray magnetic circular dichroism (XMCD) spectrum depending on the sign and magnitude of the applied voltage [4]. In the present study, we investigate the voltage-induced changes in the magnetic properties of FeCo thin films grown on the BTO substrate. Some enhancement of the voltage effect is expected owing to magnetic anisotropy of the FeCo film, which depends on the Fe-Co compositions.

II. EXPERIMENTAL

All the experiments were performed at the soft X-ray undulator beamline, BL-16A, of the Photon Factory, Japan [5]. The polarization switching between opposite circular polarizations was also applied [6, 7]. FeCo films with different Fe-Co compositions were grown on a 1 mm-thick BTO(001) substrate in a high vacuum chamber by a

simultaneous evaporation from Fe and Co rods with electron bombardment heating, keeping the substrate temperature at 373 K. A typical growth rate of the FeCo films was 0.2 nm/min. The base pressure was $\sim 3 \times 10^{-8}$ Torr, while the pressure increased to $\sim 1 \times 10^{-7}$ Torr during film deposition. Wedge-shaped samples were prepared, in which the film thicknesses were altered from 0 to ~ 8 nm. The Fe-Co compositions were chosen to be Fe₆₇Co₃₃, Fe₅₀Co₅₀, Fe₄₀Co₆₀ and Fe₂₅Co₇₅. The films were then covered with Au by electron bombardment evaporation of Au in a Mo crucible at room temperature in order to protect the films from oxidation. The voltages up to ± 1 kV were applied during the measurement between the film surface and the bottom of the substrate, which correspond to the electric fields, $E = \pm 10$ kV/cm. Here, the sign of the applied voltage is defined with respect to the bottom of the BTO substrate. It should be noted that both the electric field itself and the ferroelectric polarization of BTO substrate could affect the magnetic state of the FeCo films, and that it is difficult to separately investigate these effects because we did not perform the measurement in electric remanence of BTO. However, we assume the effect of the ferroelectric polarization, including the movement of atoms in BTO, is dominant in the present condition, since the applied electric field seems too small to directly affect the electronic structure of the FeCo films. The XMCD spectra were taken at room temperature in the grazing incidence condition, in which the angle between the surface normal and the X-ray beam was 60°, because the films show in-plane magnetization. The magnetic fields of ± 500 Oe was applied along the incident X-ray beam during the XMCD measurement. The magnetization curves were obtained by measuring the XMCD intensity at the L₃ peak top (~ 707 eV) as a function of magnetic field.

III. RESULTS AND DISCUSSION

Figure 1 displays electric field (E) dependence of magnetization curves for relatively Fe-rich samples. The magnetization curves for the Fe₆₇Co₃₃ film show a larger

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[†] Corresponding author: kenta.amemiya@kek.jp

FIG. 1. Magnetization curves for $\text{Fe}_{67}\text{Co}_{33}$ and $\text{Fe}_{50}\text{Co}_{50}$ films with ~ 2.5 nm thickness taken by applying different electric fields, E . The data for different E are intentionally shifted.

FIG. 2. Magnetization curves for $\text{Fe}_{40}\text{Co}_{60}$ and $\text{Fe}_{25}\text{Co}_{75}$ films with ~ 2.0 and ~ 2.5 nm thicknesses, respectively, taken by applying different electric fields, E . The data for different E are intentionally shifted.

hysteretic behavior, *i.e.*, enhanced hysteresis loop, at $E = \pm 5$ kV/cm compared to $E = 0$ kV/cm. This is probably due to the increase in the magnetic component which has a larger coercive field. Since BTO is known to exhibit a lattice expansion along the applied electric field, the observed voltage-induced change in the $\text{Fe}_{67}\text{Co}_{33}$ film might be attributed to the structural change, such as the formation of domain structure, of the BTO substrate, which is suggested in the 8 nm Fe film [4]. On the other hand, the magnetization curves for the $\text{Fe}_{50}\text{Co}_{50}$ film exhibit larger hysteretic behavior at positive electric fields. This could be attributed to some electronic structure effect due to the electric polarization of BTO and/or changes in the hybridization between Fe and O due to the atomic movement in BTO [8].

The magnetization curves for relatively Fe-poor samples are shown in Fig. 2. The squareness of the curve is significantly improved compared with the Fe-rich samples, probably due to stronger in-plane magnetic anisotropy of Co. On the other hand, slight voltage dependence is observed for both of the $\text{Fe}_{40}\text{Co}_{60}$ and $\text{Fe}_{25}\text{Co}_{75}$ films, even if ± 10 kV/cm electric fields are applied.

FIG. 3. XMCD intensities at the remanent state as functions of applied electric field, E , for the films with different Fe-Co compositions and thicknesses.

To quantitatively estimate the effects of the applied voltage, the XMCD intensity at the remanent state is plotted in Fig. 3 as a function of electric field, E . Remanent magnetization of the $\text{Fe}_{67}\text{Co}_{33}$ films show a minimum at $E = 0$ kV/cm, while that for the $\text{Fe}_{50}\text{Co}_{50}$ films almost monotonically increases with increasing electric field. The voltage-induced change in remanent magnetization of $\text{Fe}_{50}\text{Co}_{50}$ films between ± 5 kV/cm is $\sim 10\%$.

Figure 4 shows applied voltage dependence of the X-ray absorption and XMCD spectra for different Fe-Co compositions, taken under the magnetic field of 500 Oe. No spectral features corresponding to Fe oxides are found, which is in contrast to the previous report on pure Fe films grown on a BTO substrate [12]. We assume that the voltage-induced changes observed in the present study is not attributed to the interface oxide layer, but to the direct interaction between the BTO substrate and the FeCo films.

For the $\text{Fe}_{67}\text{Co}_{33}$ film, the XMCD intensity at the Fe L_3 peak is larger at negative voltage, which indicates a larger orbital magnetic moment in Fe [9]. Such a change in the orbital magnetic moment could be related to the change in magnetic anisotropy, but it seems too small to affect the magnetization curve in the present case. We assume that the voltage-induced change in the magnetization curve for the $\text{Fe}_{67}\text{Co}_{33}$ film is dominated by those in the substrate structure as discussed above.

For the $\text{Fe}_{50}\text{Co}_{50}$ film, a small but significant difference in the XMCD spectra are found depending on the applied voltage. In fact, the XMCD intensity is larger at both

FIG. 4. Applied voltage dependence of X-ray absorption spectra for ~ 2.5 nm $\text{Fe}_{67}\text{Co}_{33}$ film taken with opposite circular polarizations, μ^+ and μ^- , and their difference, $\mu^+ - \mu^-$ (a), and XMCD spectra for films with different Fe-Co compositions and thicknesses (b).

of the L_3 and L_2 peaks for the positive voltage, which directly indicates larger Fe magnetization. Therefore, we attribute the voltage-induced change in the magnetization curve of the $\text{Fe}_{50}\text{Co}_{50}$ film to that in Fe magnetization at the saturation state. It may seem strange that the difference in the XMCD intensities between $E = \pm 5$ kV/cm is relatively large at remanence (Fig. 3), while that at saturation is rather small (Fig. 4b). However, since the hysteresis curve of the $\text{Fe}_{50}\text{Co}_{50}$ film is far from square, the remanent magnetization can be quite sensitive to, e.g., coercivity of the film. In fact, the coercive field of the $\text{Fe}_{50}\text{Co}_{50}$ film at $E = +5$ kV/cm is much larger than those at other electric fields.

In contrast, no significant voltage dependence is found in the XMCD spectra for $\text{Fe}_{40}\text{Co}_{60}$ and $\text{Fe}_{25}\text{Co}_{75}$. We would just notify that the positive shoulder-like structure at ~ 710 eV in the XMCD spectra decreases as the Co composition increases, which could be interpreted as the change in the diffuse magnetic moment in Fe [10].

Finally, let us discuss the peculiar behavior of the $\text{Fe}_{50}\text{Co}_{50}$ films, by referring to a self-consistent local spin-density functional calculation for FeCo alloys [11]. It was reported that the density of state (DOS) for the majority spin band has a broad shoulder around the Fermi level, whereas the Fermi level is located in a valley of DOS for the minority band, in the case of pure Fe. On the other hand, DOS for the majority band of the $\text{Fe}_{50}\text{Co}_{50}$ alloy is quite small and flat around the Fermi level, whereas that for the minority band exhibits an abrupt change at the Fermi level, especially if the alloy takes the CsCl structure. Therefore, it is expected that the magnetic properties of the $\text{Fe}_{50}\text{Co}_{50}$ alloy is sensitive to a subtle change in

the electronic structure induced by the applied voltage.

IV. CONCLUSION

We have investigated the effects of applied voltage on the magnetic properties of FeCo thin films with different Fe-Co compositions grown on a BaTiO_3 substrate. The magnetization curve for the $\text{Fe}_{67}\text{Co}_{33}$ film show a minimum hysteretic behavior at zero voltage, whereas the $\text{Fe}_{50}\text{Co}_{50}$ film exhibits larger hysteretic behavior at positive voltages. On the other hand, the $\text{Fe}_{40}\text{Co}_{60}$ and $\text{Fe}_{25}\text{Co}_{75}$ films show almost no dependence on the applied voltage. The XMCD spectra suggest that magnetization of the $\text{Fe}_{50}\text{Co}_{50}$ film at the saturation state is larger at the positive applied voltage, while those for other compositions have no detectable dependence on the applied voltage. The peculiar behavior for $\text{Fe}_{50}\text{Co}_{50}$ could be attributed to the abrupt change in the density of state at the Fermi level.

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