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Voltage-dependent magnetic properties of Co/$BiFeO_3$ heterostructure: x-ray magnetic circular dichroism study

Zhendong CHI[1,*], Hiroyuki YAMADA[2], Goro SHIBATA[1], Shoya SAKAMOTO[1], Yosuke NONAKA[1], Keisuke IKEDA[1], Masahiro SUZUKI[1], Yuxuan WAN[1], Masako SAKAMAKI[3], Kenta AMEMIYA[3], Akihito SAWA[2] and Atsushi FUJIMORI[1]

[1]Department of Physics, The University of Tokyo, Bunkyo, Tokyo, 113-0033, Japan
[2]National Institute of Advanced Industrial Science and Technology, Tsukuba, 305-8565, Japan
[3]Photon Factory, 1-1 Oho, Tsukuba, 305-0801, Japan

1 Introduction

Aiming to realize low-energy consumption through the application of spintronics, i.e., controlling of spin states by electric current or voltage, a variety of methods have been proposed using spin accumulation or spin transfer by electric current, e.g., current-induced domain-wall motion and magnetization reversal [1,2] and current control of magnetization in ferromagnetic semiconductors [3]. However, the current densities required to transfer the spin moment in such systems are very large, still resulting in a large loss of energy. Another approach to control magnetization is to use electric voltage. A promising series of materials in the application of voltage control of magnetization are magnetoelectric multiferroics [4,5]. Bismuth ferrite ($BiFeO_3$, BFO) is the most studied multiferroic material as it is the only material showing multiferroic properties at room temperature so far. BFO has a nearly G-type antiferromagnetic order with finite incommensurability and the electrical polarization of BFO is along the (111) orientation. Due to the spin canting induced by Dzyaloshinskii-Moriya interaction [6,7], weak ferromagnetism also exists in BFO [8] and its orientation can be modulated by an electric field [9].

Because of the weak magnetization of BFO, ferromagnet (FM)/BFO thin film heterostructures are often used to realize the voltage control of magnetization in such systems. As usual, oxides, e.g., CrO_2, $La_{0.7}Sr_{0.3}MnO_3$, and $La_{0.5}Ca_{0.5}MnO_3$, are used for the FM layer [10-13]. However, the T_c of the magnetic oxide is often lower than room temperature, which prohibits their practical applications. By domain architecture utilizing the strain of substrates, it has been found that stripe-like ferroelectric domains of BFO can be formed on a (110)-oriented $DyScO_3$ (DSO) substrate [14]. Then by depositing CoFe alloy onto the BFO thin film with the magnetic domain of the same pattern, the 180° reversal of magnetization at room temperature has been realized [15].

Though the realization of electrical-field control of magnetization in CoFe/BFO heterostructure at room temperature, the nature of the magnetic coupling mechanism between the metallic alloy CoFe and the perovskite oxide BFO is still unknown. It has been known that the polarization and the Dzyaloshinskii-Moriya vector of BFO can be reversed after applying a voltage along the c-axis, resulting in the 180° reversal of weak moment of Fe [9]. It is theoretically shown that the magnetic moment of Fe in BFO is enhanced at the CoFe/BFO interface and coupled with the magnetic moment of CoFe [16,17]. As a result, the magnetic moment of CoFe should be also varied when modulating the magnetic moment of Fe in BFO. However, direct evidences for the magnetic moment of Fe in BFO and the coupling mechanism between BFO and CoFe have still remained uncertain so far. In this report, by using element-specific x-ray magnetic circular dichroism (XMCD), the magnetic properties and their voltage dependence of Fe and Co in a Co/BFO heterostructure has been studied. The magnetic moment of Fe in BFO at room temperature was confirmed by XMCD. Furthermore, the magnetic moments of both Co and Fe have also been found to vary under different electric voltages.

2 Experiment

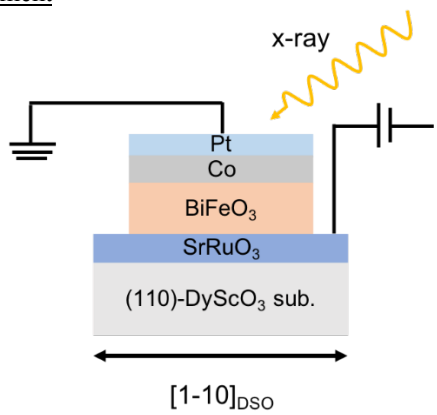

Figure 1. Setup for the voltage-dependent XAS and XMCD measurements.

An 80 nm BFO (001)/$SrRuO_3$ (SRO) thin film was grown on an (110)-oriented DSO substrate by the pulsed laser deposition (PLD) method. SRO thin film was used as an electrode to apply voltage. 1nm Co was deposited on BFO by electron-beam evaporation method, followed by a 2 nm Pt thin film used as a capping layer and an electrode. X-ray absorption spectroscopy (XAS) and XMCD measurements were performed at the helical undulator beamline BL-16A1 of KEK-PF. The spectra were collected both in the total electron yield (TEY) mode and the total

fluorescence mode (TFY). The measurements were all performed at room temperature in an ultrahigh vacuum below 4×10^{-7} Torr. The magnetic field was set at 1.2 T and parallel to the incident x rays. There was an angle of 60° between the magnetic field and the sample normal during the measurement. The projection of the magnetic field on the ab plane of the film was along the (1-10) direction of the DSO substrate. A voltage was applied between Pt and SRO before each measurement. The experimental setup is illustrated in Fig. 1.

3 Results and Discussion

Figure 2 shows the XMCD spectra at the Fe and Co $L_{3,2}$-edges of Co/BFO heterostructure measured at room temperature. In Fig. 2(a)-(c), it can be found that there are clear XMCD signals of Fe after adding different external voltages. The spectral line shapes are similar to what have been observed on the ferromagnetic oxide/BFO interface at low temperature so far [12,13]. From Fig. 2(a)-(c), one finds that the XMCD intensity changed after the poling with different voltage direction. Although the XMCD spectra under different orientations of magnetic field show different intensities, we note that the background is rather strong here and further studies are needed to confirm the differences. Figure 2(d)-(f) illustrate the XMCD spectra of Co in Co/BFO heterostructure under different directions of magnetic field and after different poling voltages and directions. For comparison of the XMCD intensity, the XMCD spectra measured under a -500 Oe magnetic field have been reversed in all the three panels. In all the XAS spectra of Co, a shoulder exists on the high energy side, suggesting a partially oxidized state of Co in the heterostructure. Similar to the results of the Fe $L_{3,2}$ edge, the XMCD intensities of Co are also asymmetric and changed after the poling of voltage with orientation reversal.

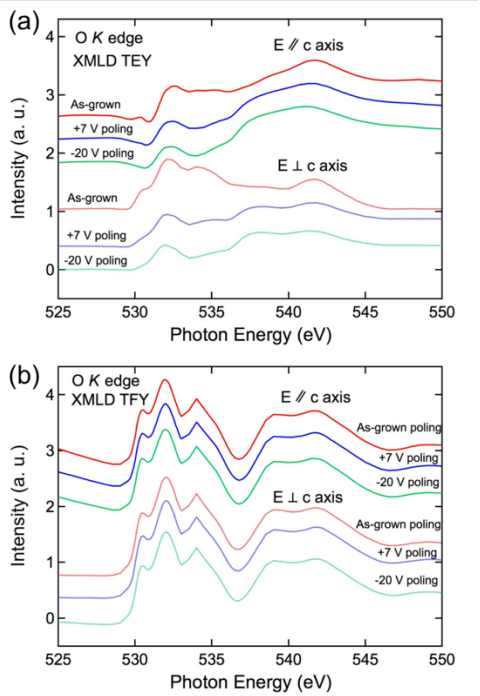

Figure 3. XAS spectra at the O *K* edge collected in the TEY mode (a) and TFY mode (b).

In order to understand the coupling mechanism between Co and BFO, XAS at the oxygen *K* edge was also measured using x rays with linear polarization. Figures 3(a) and (b) illustrate the O *K*-edge XAS collected using the TEY and TFY modes, respectively. The line shape of O *K*-edge XAS after the poling is much different from that at the as-grown

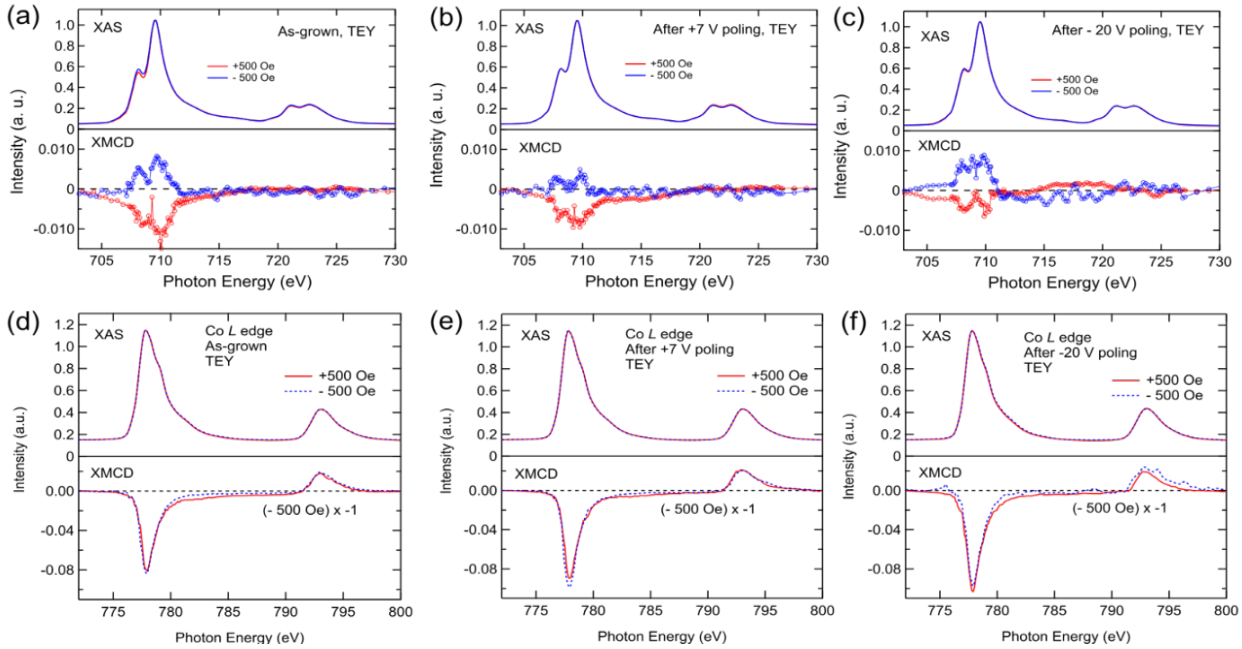

Figure 2. XAS and XMCD spectra at the Fe and Co $L_{3,2}$-edges of Co/BFO heterostructure. (a) Fe $L_{3,2}$ edges at the as-grown state, (b) After a +7 V voltage poling, (c) After -20 V voltage poling. (b)-(d) Co $L_{3,2}$ edges under the same condition as Fe.

state in the TEY mode but is almost the same in the TFY mode. This may be due to the diffusion of oxygen atoms across the Co/BFO interface. However, we cannot exclude the possibility of the removal of adsorbed oxygen atoms on the surface of the sample.

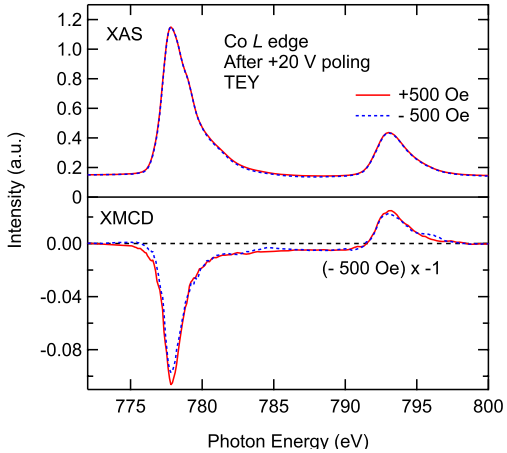

Figure 4. XAS and XMCD spectra at the Co $L_{3,2}$ edge after a +20 V voltage poling.

After the -20 V poling, another +20 V voltage was then applied to the Co/BFO heterostructure to check the reproducibility of the voltage dependence. XAS and XMCD spectra at the Co $L_{3,2}$ edges are illustrated in Fig. 4. The asymmetry of the XMCD intensity under the magnetic field reversal is the same as that after the -20 V poling. This may indicate that the BFO thin film was electrically broken down after the application of the too high voltage of the first -20 V poling. Although the reproducibility of the voltage dependence could not be confirmed, our results still provide direct evidence for the existence of the magnetic moment of Fe in BFO at room temperature and there should be a magnetic coupling between Co metal and Fe in BFO at the Co/BFO interface. Further studies are needed to confirm the magnetic field and voltage dependence of Fe and Co $L_{3,2}$-edges XMCD and investigate the coupling mechanism of Co and Fe at the Co/BFO interface.

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* chi@wyvern.phys.s.u-tokyo.ac.jp