Effect of Electric Field on Magnetism of Ni Thin Films via Antiferromagnetic NiO

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Effects of electric fields on magnetization of ferromagnetic Ni films grown on a Cu(001) single crystal covered with antiferromagnetic NiO overlayer are investigated by means of X-ray absorption spectroscopy (XAS), X-ray magnetic circular dichroism (XMCD), and magneto-optical Kerr effect (MOKE). The growth of the NiO overlayer on the Ni film is confirmed by XAS, and it is revealed by XMCD and MOKE that the NiO/Ni films show a spin reorientation transition from in-plane to perpendicular magnetization with increasing Ni thickness. It is also observed that the coercive field of the Ni films increases as the NiO thickness increases, possibly due to the interaction with antiferromagnetic NiO. Remanent magnetization of the Ni film is found to be modified by the application of electric fields. The possible origin of the electric-field effects is discussed, and some change in magnetic anisotropy of the Ni film is suggested. [DOI: 10.1380/ejssnt.2018.186]

Keywords: Field effect; Magnetic interfaces; Ni; NiO; Cu(001)

I. INTRODUCTION

The control of magnetism by the electric field 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 demonstrated for FeCo thin films [1]. The idea to control magnetization by the electric field has been proposed as a multiferroic concept, and has been realized, for instance, in BiFeO3, which is one of the most typical multiferroic materials. On the other hand, it was reported that the coercive field of a ferromagnetic Fe thin film grown on a ferroelectric BaTiO3 (BTO) substrate is modified by voltage application between the Fe film and the bottom of the BTO substrate [2]. Moreover, we have recently investigated the electric field-induced changes in magnetic properties for Fe/BTO by means of X-ray absorption spectroscopy (XAS) [3]. It has been revealed that a few nm-thick Fe oxide layer exists at the Fe/BTO interface, and that it plays an essential role in the electric-field effects, which suggests that a ferromagnetic/antiferromagnetic coupling could be altered by electric fields.

NiO is one of the most typical antiferromagnetic insulators, while Ni is a ferromagnetic metal. Therefore, both of the ferromagnetic/antiferromagnetic and metal/insulator interactions are expected at the Ni/NiO interface, which gives a possibility to control the magnetic properties on Ni by the electric field. Moreover, Ni thin films grown on a Cu(001) single crystal have been well known to exhibit peculiar magnetic anisotropy, i.e., spin reorientation transitions (SRTs); in-plane, perpendicular, and again in-plane magnetization with increasing Ni thickness [4, 5].

In the present study, we investigate the effects of the electric field on the magnetic properties of NiO/Ni films grown on a Cu(001) substrate, by means of XAS, X-ray magnetic circular dichroism (XMCD) and magneto-optical Kerr effect (MOKE).

II. EXPERIMENTAL

A Cu(001) single crystal was cleaned by repeated cycles of Ar+ sputtering at 1.5 kV and annealing at ~900 K. The substrate was first exposed to 5 × 10⁻⁴ Pa oxygen for 300 s at 500 K, which leads to single-layer atomic oxygen adsorption on Cu(001). Co and Ni films were subsequently grown on the O/Cu(001) surface at room temperature by the electron-bombardment heating of Co and Ni rods. Here, very thin Co layer was inserted between the Ni films and the substrate in order to increase the critical thickness for the SRT of Ni. During the Co and Ni deposition, oxygen remains at the surface, resulting in O/Ni/Cu/Cu(001) [6, 7]. NiO overlayers were then grown by a reactive deposition of Ni in 1 × 10⁻³ Pa oxygen, and NiO/Ni/Co/Cu(001) films were prepared. For the experiment with electric-field application, the films were further covered with a SiO₂ insulating layer, which was deposited by the electron-bombardment heating of SiO₂ powders in a Ta crucible. Finally, a Au electrode was deposited by the electron-bombardment heating of Au wires in a Ta crucible. Voltages were applied between the Au electrode and the Cu(001) substrate, and the electric potential at the substrate was defined to be 0 V.

For the characterization of the films, Ni K-edge XAS was measured at BL-12C in the Photon Factory, High Energy Accelerator Research Organization (KEK-PF) in the fluorescence-yield mode with a 19-element solid-state detector. Ni L-edge XAS and XMCD data were recorded at BL-7A and BL-16A at KEK-PF [8-10] in the total-electron-yield mode with a drain current and the total-fluorescence-yield mode with a microchannel-plate detector. MOKE measurements were performed by using a 670 nm semiconductor laser. All of the XAS, XMCD, and MOKE measurements were performed at room temperature.

III. RESULTS AND DISCUSSION

Figure 1 shows Ni K-edge XAS data, from which chemical states of the films are investigated. The obtained
spectra systematically change depending on the Ni thickness, and it is clearly recognized that a shoulder structure, marked as A, increases, while the main peak, indicated by B, decreases with increasing Ni thickness. Since A and B are typical spectral features for Ni metal and NiO, respectively, it is confirmed that the NiO/Ni bilayers were prepared as expected.

To further examine the film configuration, a series of Ni L-edge XAS is shown in Fig. 2 for different Ni thicknesses. Since the obtained spectra consist of Ni and NiO XAS components, one can estimate the configuration of the NiO overlayer from the Ni-thickness dependence of the spectra. The spectral component for metallic Ni is plotted in Fig. 2, which is estimated from least-square fitting for each spectrum by using the XAS for NiO (0 nm)/Ni (2 nm) and NiO (0.9 nm)/Ni (0 nm) films as standards for Ni and NiO, respectively. Note that the signal from the Ni layer does not increase linearly with Ni thickness, because the emitted electrons after the X-ray absorption decay exponentially when they go through the Ni and NiO layers. The decay factor of the emitted electrons is given by \( \exp(-z/\lambda) \), where \( z \) and \( \lambda \) represent the depth at which the electrons are created and probing depth, respectively. The NiO thickness is then estimated to be 0.95 nm, by fitting the Ni-thickness dependence of the metallic Ni component in XAS, assuming that the probing depth in the total-electron-yield mode is 3.5 nm. The estimated NiO thickness is close to the designed value (0.9 nm), indicating that the NiO layers grow on the Ni films as expected. Some deviation from the fitted curve is recognized, however, which might suggest intermixing between the Ni and NiO layers.

A typical Ni L-edge XMCD spectrum is given in Fig. 3. The X-ray absorption spectra, \( \mu^+ \) and \( \mu^- \), show several peaks which are characteristic to Ni and NiO, while the XMCD difference spectrum is similar to that for the Ni metal. This directly indicates that the NiO overlayer has no net magnetization, which is quite reasonable because NiO is an antiferromagnetic material.

Figure 4 shows Ni-thickness dependence of the magnetization curve, which was obtained by XMCD measurements. Note here that the XMCD data were obtained in the total-fluorescence-yield mode. Applied magnetic field was 500 Oe, which is parallel to the incident X-ray beam and thus perpendicular to the surface.
at 4 nm Ni thickness, while in-plane magnetic anisotropy is suggested for 2.5 nm Ni. This result is consistent with the previous reports [4, 5], which indicated a SRT from in-plane to perpendicular magnetization with increasing Ni thickness, though the critical thickness for the SRT is much thicker for the present case, due to in-plane magnetic anisotropy of the Co layers as expected. On the other hand, NiO-thickness dependence of magnetization curve is shown in Fig. 5, which was obtained by polar MOKE measurements. The coercive field significantly increases at 1 nm NiO, as shown in Fig. 5, which is attributed to the appearance of the antiferromagnetic character of the NiO layer. Thus, it is suggested that Néel temperature, $T_N$, of 1 nm NiO is close to room temperature, which is reasonable considering that $T_N$ for bulk NiO is $\sim 500$ K.

Finally, the effects of the electric fields are investigated. Figure 6 shows magnetization curves taken with applied voltages of $+10$ V, $0$ V, and $-10$ V. An increase in remanent magnetization is found for the negative voltage, as also shown in Fig 6, though the experimental errors are rather large. One may attribute the observed electric-field dependence to a change in the antiferromagnetic character of NiO due to, e.g., an increase in the NiO thickness induced by the redox reaction at the Ni/NiO interface. However, electric-field dependence of the coercive field is quite small as seen in Fig. 6, which indicates little change in the antiferromagnetic character of the NiO layer. On the other hand, the magnetization curve at $+10$ V is slightly inclined, which suggests that the perpendicular magnetic anisotropy of the film is suppressed compared to that at $-10$ V. Thus, we suppose the origin of the electric-field dependence of Ni magnetization might be some change in magnetic anisotropy of the Ni films, which has been also reported for Fe$_{80}$Co$_{20}$ films [11]. Although the electric field-induced modification in magnetic anisotropy via structural change of the substrate was observed in Cu/Ni multilayers grown on a BTO substrate [12], it does not seem to fit the present case, because it is unlikely that the structure of NiO and/or SiO$_2$ layers are modulated by the electric field. Further studies are necessary to achieve electric-field control of magnetic properties, especially via the antiferromagnetic-ferromagnetic interaction between NiO and Ni, by optimizing, e.g., the Ni and NiO thicknesses.

IV. CONCLUSION

Effects of electric fields on magnetization of ferromagnetic Ni films grown on a Cu(001) single crystal covered with antiferromagnetic NiO overlayers have been investigated by means of XAS, XMCD, and MOKE. The growth of the NiO overlayer by reactive deposition on the Ni film was confirmed. It was revealed that the NiO/Ni films show a spin reorientation transition from in-plane to perpendicular magnetization with increasing Ni thickness, and that the coercive field of the Ni film increases as the NiO thickness increases due to the appearance of the antiferromagnetic character of NiO. It was found that remanent magnetization of the film is modified by the application of electric fields. The possible origin of the electric-field effects was discussed, and some change in magnetic anisotropy of the Ni film was suggested.

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[1] Y. Shiota, T. Nozaki, F. Bonell, S. Murakami, T. Shinjo, and Y. Suzuki, Nat. Mater. 11, 39 (2012).
[2] S. Brivio, D. Petti, R. Bertacco, and J. C. Cezar, Appl. Phys. Lett. 98, 092505 (2011).
[3] M. Sakamaki and K. Amemiya, e-J. Surf. Sci. Nanotech. 13, 139 (2015).
[4] B. Schulz and K. Baberschke, Phys. Rev. B 50, 13467 (1994).
[5] W. L. O’Brien, T. Droubay, and B. P. Tonner, Phys. Rev. B 54, 9297 (1996).
[6] R. Nünthel, T. Gleitsmann, P. Poulopoulos, A. Scherz, J. Linder, E. Kosubek, Ch. Litwinski, Z. Li, H. Wende, K. Baberschke, S. Stolbov, and T. S. Rahman, Surf. Sci. 531, 53 (2003).
[7] K. Amemiya and M. Sakamaki, Appl. Phys. Lett. 98, 012501 (2011).

[8] K. Amemiya, H. Kondo, T. Yokoyama, and T. Ohta, J. Electron Spectrosc. Relat. Phenom. 124, 151 (2002).
[9] K. Amemiya, A. Toyoshima, T. Kikuchi, T. Kosuge, K. Nigorikawa, R. Sumii, and K. Ito, AIP Conf. Proc. 1234, 295 (2010).
[10] K. Amemiya, M. Sakamaki, T. Koide, K. Ito, K. Tsujiya, K. Harada, T. Aoto, T. Shioya, T. Obina, S. Yamamoto, and Y. Kobayashi, J. Phys.: Conf. Ser. 425, 152015 (2013).
[11] Y. Shiota, T. Maruyama, T. Nozaki, T. Shinjo, M. Shiraiishi, and Y. Suzuki, Appl. Phys. Express 2, 063001 (2009).
[12] Y. Shirahata, R. Shiina, D. L. Gonzalez, K. J. A. Franke, E. Wada, M. Itoh, N. A. Pertsev, S. van Dijken, and T. Taniyama, NPG Asia Mater. 7, e198 (2015).