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Manipulation of magnetic properties of ferromagnetic Ni thin films grown on Cu(001) by antiferromagnetic NiO and effects of voltage application

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The magnetic properties of ferromagnetic Ni thin films grown on a Cu(001) single crystal are modified by the growth of a NiO overlayer, as well as by the voltage application through the NiO/Ni interface. A spin reorientation transition from in-plane to perpendicular magnetization is induced with increasing NiO thickness, and the coercive field significantly increases by further growth of the NiO overlayer. The remanent magnetization of the films is found to be modulated by the voltage application. Moreover, a small exchange-bias effect arising from the ferromagnetic–antiferromagnetic interaction at the interface is observed, and the amplitude of the effect is modified by the applied voltage.

1. Introduction

The manipulation of the magnetic properties of thin films such as magnetic anisotropy has attracted much interest in the last few decades. For instance, 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.[1,2] Moreover, the modification of magnetic anisotropy of the Ni/Cu(001) films by the adsorption of H_2, CO, and O has been reported,[3–5] as well as by the deposition of Cu, Fe, and FeMn.[4,6–8]

Recently, it has been reported that the magnetic anisotropy of $Fe_{80}Co_{20}$ films is modified by the voltage application.[9] It was also reported that the coercive field of ferromagnetic Fe thin films grown on a ferroelectric $BaTiO_3$ (BTO) substrate is modified by the voltage application.[10] Moreover, we have recently investigated the effects of the voltage on the magnetic properties of Fe/BTO films, and revealed that a 1–2-nm-thick Fe oxide layer exists at the Fe/BTO interface, which plays an essential role in the voltage effects.[11] Since most Fe oxides are antiferromagnetic or ferrimagnetic materials, which contain some antiferromagnetic features, it is suggested that the ferromagnetic/antiferromagnetic interaction could be modified by the voltage application.

In this study, we investigate the effects of NiO overlayers on the magnetic properties, especially magnetic anisotropy, of Ni/Cu(001) films, and those of the voltage application, by means of X-ray magnetic circular dichroism (XMCD) and the magneto-optical Kerr effect (MOKE). Since NiO is one of the most typical antiferromagnetic insulators, while Ni is a ferromagnetic metal, a modification of the ferromagnetic/antiferromagnetic interaction by the voltage application at the Ni/NiO interface is expected.

2. Experimental procedure

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). Ni films were then grown on the O/Cu(001) surface at room temperature by the electron-bombardment heating of a Ni rod. During the Ni deposition, oxygen remains at the surface, resulting in O/Ni/Cu(001).[12,13] NiO overlayers were grown by a reactive deposition of Ni in 1×10^{-4} Pa oxygen, and NiO/Ni/Cu(001) films were prepared. In some cases, a very thin Co layer was inserted between the Ni films and the Cu substrate, in order to increase the critical thickness of Ni films for the SRT to perpendicular magnetization. For the experiments with voltage application, the films were further covered with a SiO_2 insulating layer, which was deposited by the electron-bombardment heating of SiO_2 powders in a Mo crucible. Finally, a Au electrode was deposited by the electron-bombardment heating of Au wires in a Mo 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.

Ni L-edge XMCD data were recorded at the soft X-ray beamline BL-16A in the Photon Factory, High Energy Accelerator Research Organization.[14,15] The total-electron-yield mode with a drain current measurement was adopted, and the XMCD spectra was obtained in the normal-incidence configuration, which detects the perpendicular component of Ni magnetization. Magnetic fields of ± 1000 Oe were applied before each measurement, and remanent magnetization was observed. The direction of the applied magnetic field was parallel to the incident beam, which was thus perpendicular to the sample surface. Polar MOKE measurements, which also detect perpendicular magnetization, were performed by using a 670 nm semiconductor laser. Both the XMCD and MOKE data were obtained at room temperature.

3. Results and discussion

Figure 1 shows a typical XMCD spectrum, which was taken for NiO (1 nm)/Ni (1.6 nm)/Cu(001). Some spectral features characteristic to NiO are found in the absorption spectra, μ^+ and μ^-, which are indicated by arrows. On the other hand, the XMCD difference spectrum, $\mu^+ - \mu^-$, exhibits a typical spectral shape for the Ni metal,[6,8,16] which indicates that the magnetization of the film originates from the Ni layer. Since the data were recorded for the remanent state in the normal-incidence configuration, which detects perpendicular magnetization, it is directly revealed that the film has perpendicular magnetic anisotropy.

The NiO thickness dependence of the XMCD spectra is shown in Fig. 2. The absorption spectrum for the film

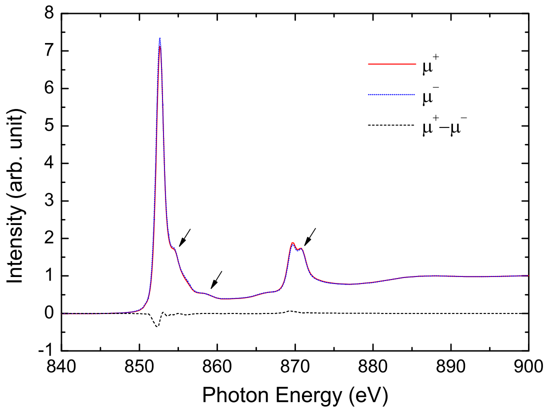

Fig. 1. (Color online) Normal-incidence Ni L-edge XMCD spectrum for NiO (1 nm)/Ni (1.6 nm)/Cu(001) taken at the remanent state.

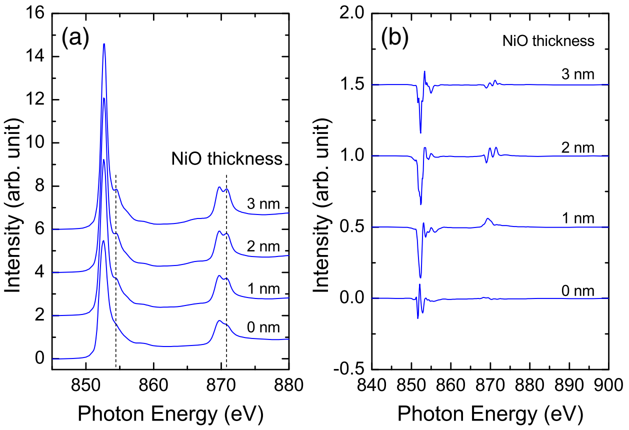

Fig. 2. (Color online) Normal-incidence Ni L-edge polarization averaged (a) and XMCD difference (b) spectra for NiO (x nm)/Ni (1.6 nm)/Cu(001) taken at the remanent state.

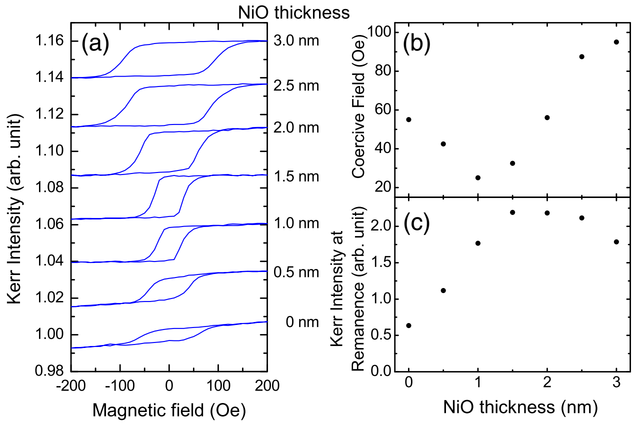

Fig. 3. (Color online) Magnetization curve for Au (5 nm)/ SiO_2 (300 nm)/NiO (x nm)/Ni (1.6 nm)/Cu(001) obtained by polar MOKE measurement (a), and coercive field (b) and Kerr intensity at the remanent state (c) as functions of NiO thickness.

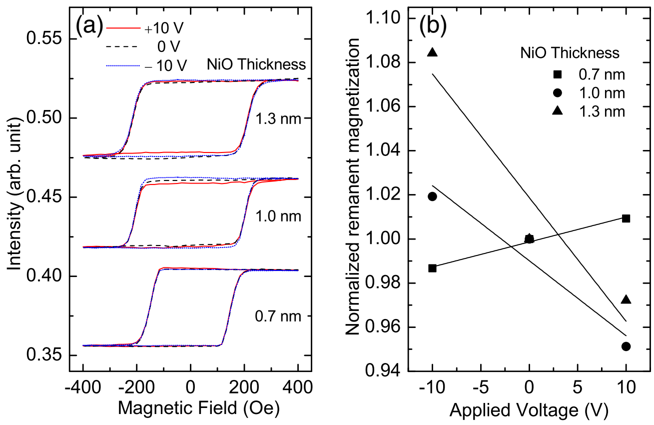

Fig. 4. (Color online) Applied voltage dependence of magnetization curve for Au (5 nm)/SiO_2 (100 nm)/NiO (x nm)/Ni (3 nm)/Co (0.2 nm)/ Cu(001) obtained by polar MOKE measurement (a) and normalized remanent magnetization as a function of applied voltage (b).

without Ni corresponds to that for O/Ni/Cu(001), and is similar to that shown in a previous report.[13] As the NiO thickness increases, it is recognized that the spectral features of NiO in the absorption spectra are more prominent for thicker NiO, as indicated by dashed lines. On the other hand, although the XMCD spectra are small and noisy owing to the relatively sharp spectral features for NiO, which do not exhibit XMCD, the XMCD intensity is maximum at 1–2 nm NiO thickness and decreases with thicker NiO. The thickness dependence of XMCD intensity is interpreted by assuming that the Ni films show no or little perpendicular magnetization without NiO, and then exhibit a SRT to perpendicular magnetization by deposition of ~1–2 nm NiO. In that case, the XMCD intensity decreases with thicker NiO even if perpendicular magnetization in the Ni film is kept constant, because the XMCD intensity reflects the amplitude of perpendicular magnetization per Ni atom in both the Ni and NiO layers.

The NiO-overlayer-induced SRT is further confirmed by the polar MOKE data shown in Fig. 3. The magnetization curve at 0 nm NiO is inclined and shows little remanent magnetization, which suggests the in-plane magnetic anisotropy of the film. As the NiO thickness increases, the remanent magnetization increases and almost saturates at 1 nm NiO, indicating the SRT to perpendicular magnetization. Moreover, the coercive field of the film significantly increases at thicker NiO, which can be attributed to the appearance of the antiferromagnetic feature of NiO. Thus, it is revealed that the NiO overlayers have two effects on the magnetization of Ni films: enhancement of perpendicular magnetic anisotropy and increase in coercive fields. One can also find some decrease in the MOKE signal for 3 nm NiO, which might be another effect of the antiferromagnetic NiO layer. Further experiments for thicker NiO films are necessary to quantitatively pursue such effect.

Figure 4 shows the applied voltage dependence of the magnetization curves of the films with relatively thin NiO layers, in which the NiO layers are around the phase transition to the antiferromagnetic state. It is found that the remanent magnetization is reduced and the magnetization curve is inclined for a positive voltage at NiO thicknesses of 1.0 and 1.3 nm. Such voltage dependence can be attributed to the decrease in perpendicular magnetic anisotropy of the Ni film by applying a positive voltage, as is the case with, for example, $Fe_{80}Co_{20}$.[9] It is interesting that an opposite voltage dependence is found at 0.7 nm NiO, although the effect is relatively small. Since the 0.7 nm NiO film is supposed to be too thin to show an antiferromagnetic feature as discussed

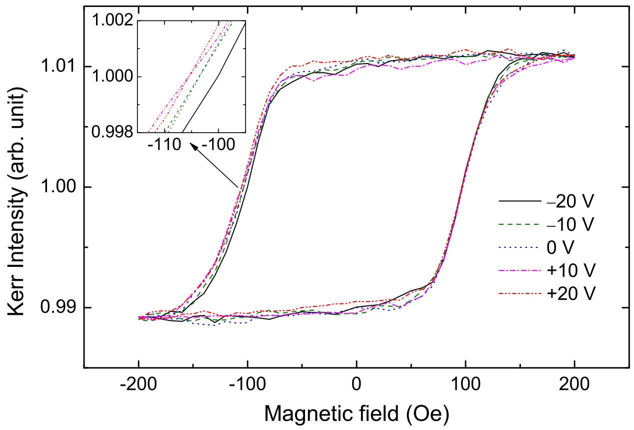

Fig. 5. (Color online) Applied voltage dependence of magnetization curve for Au (5 nm)/SiO_2 (300 nm)/NiO (2.5 nm)/Ni (1.6 nm)/Cu(001) obtained by polar MOKE measurement.

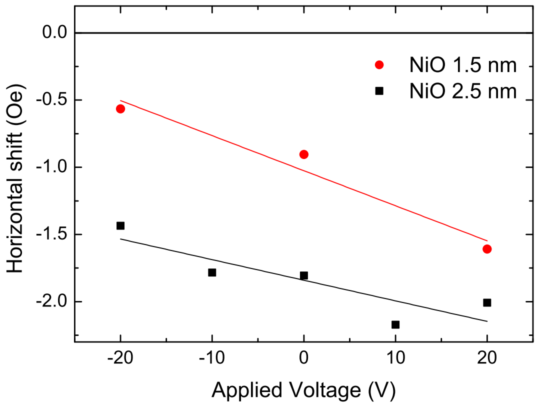

Fig. 6. (Color online) Horizontal shift of magnetization curve for Au (5 nm)/SiO_2 (300 nm)/NiO (x nm)/Ni (1.6 nm)/Cu(001) as a function of applied voltage.

above, the difference in the effect of the applied voltage between the 0.7 nm NiO overlayer and thicker ones might be attributed to the difference in the magnetic structure of NiO.

For thicker NiO layers, horizontal shifts in the magnetization curves, as well as the voltage dependence of the shifts, are observed as shown in Figs. 5 and 6. The horizontal shift in the magnetization curve itself is interpreted as the exchange-bias effect arising from the magnetic interaction between ferromagnetic Ni and antiferromagnetic NiO layers. One may suppose that the voltage dependence is attributed to the change in the NiO thickness via the redox reaction at the NiO/Ni interface, which has been observed for GdO_x/Co.[17] However, the NiO thickness should increase when a negative voltage is applied, if the redox reaction is induced by the oxygen-ion migration. This leads to the increase in the exchange bias effect at a negative applied voltage, which is opposite to the observed results. On the other hand, the electric-field control of the exchange bias effect has been reported by using the multiferroic nature of $BiFeO_3$[18,19] or electric-field-induced strain in ferroelectric substrates,[20,21] both of which are in different situations from the present case, i.e., no intrinsic multiferroic material is included in the present samples, and NiO is a dielectric material. Therefore, another origin of the voltage dependence of the exchange-bias effect has to be considered.

We tentatively speculate that the voltage application to the NiO/Ni interface induces changes in the magnetic anisotropy of Ni, and possibly of NiO, which results in the modulation in the exchange interaction between NiO and Ni. In fact, the electric-field-induced changes in magnetic anisotropy at the interface between ferromagnetic thin films and dielectric MgO have been frequently reported in this decade.[9,22–28] Further experimental and theoretical studies are required to clarify the origin of the observed voltage-dependent changes in the exchange-bias effect.

4. Conclusions

The magnetic properties of NiO/Ni thin films grown on a Cu(001) single crystal and the effects of the voltage application through the NiO/Ni interface have been investigated by means of XMCD and MOKE. It was revealed that the spin reorientation transition from in-plane to perpendicular magnetization is induced with increasing NiO thickness, and the coercive field significantly increases by the further growth of the NiO overlayer. The remanent magnetization of the films was found to be modulated by the voltage application, which can be attributed to the changes in the magnetic anisotropy of the Ni films. The exchange-bias effect was observed, which arises from the ferromagnetic–antiferromagnetic interaction at the interface. Moreover, it was revealed that the amplitude of the exchange-bias effect is modified by the voltage application, which might be explained by the voltage-induced modulation of the exchange interaction between the Ni and NiO layers.

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