Temperature dependence of remanent magnetization of thin films at the interface to a nonmagnetic material: Cu/Ni/Cu(100)

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Temperature dependence of remanent magnetization of Ni thin films at the interface to Cu is investigated by means of the depth-resolved x-ray magnetic circular dichroism technique. Although magnetization at the interface is smaller than that in the inner layers, no detectable difference in Curie temperature, at which remanent magnetization vanishes, is found between the interface and the inner layers. In contrast, the magnetization-temperature curves at the interface and in the inner layers show a small difference, which cannot be explained by using a simple mean-field theory if the interface layer is assumed to have the same exchange-coupling constant as that in the inner layers. It is also shown that the critical exponent for the in-plane magnetized film is significantly larger than that for the perpendicularly magnetized film due to the difference in the degree of freedom for the magnetic moment.

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I. INTRODUCTION

The magnetic properties of ferromagnetic ultrathin films, such as Ni and Co, especially the critical phenomena around Curie temperature T_C, have been extensively investigated in past decades.[1–10] For a particular system, Ni/Cu(100), T_C has been reported to decrease from ∼540 K for 16 monolayers (MLs) to ∼200 K for 3 MLs,[3] whereas, it is 627 K for bulk Ni. More interestingly, the critical exponents for Ni films thicker than 10 MLs and thinner than 8 MLs were reported to be ∼0.4 and ∼0.23, respectively, which are attributed to the crossover from the three-dimensional Heisenberg to finite-size two-dimensional behavior.[3,10] These results are deduced by layer-averaged measurements, however, so that no information on the surface layer has been obtained. Although it is expected that T_C at the surface or interface is the same as that in the inner layers as far as the film has no carrier density gradient, here, we report experimental evidence for this fundamental problem. Moreover, it must be interesting how magnetization at the surface or interface behaves upon approaching T_C.

Some theoretical studies have been reported in order to clarify the layer-resolved behavior of magnetization as a function of temperature.[4,7,9] Jensen and Bennemann[4] adopted a simple mean-field theory in which the exchange interaction constant J at the interface is assumed to be the same as that in the inner layers. Busiakiewicz *et al.*[9] showed layer-resolved magnetization-temperature curves by means of the effective field theory. They adopted different J's at the interface and in the inner layers, $J = 1.7 \times 10^{-21}$ and 4.97×10^{-22} J, respectively, which had been reported by Scherz *et al.*[8] Herrmann *et al.*[7] applied the Hubbard model with a generalization of the spectral-density approach. All of the above-mentioned theoretical studies showed that magnetization at the surface or interface to the nonmagnetic layer is smaller than that in the inner layers. It has also been indicated that T_C at the surface or interface is the same as that in the inner layers. No experimental data have been reported, however, which evidence such behavior at the surface or interface.

In the present study, we separately determine remanent magnetization of Ni thin films at the interface to nonmagnetic Cu and in the inner Ni layers by means of the depth-resolved x-ray magnetic circular dichroism (XMCD) technique[11–16] by changing the Ni film thickness, d_{Ni}, and the temperature. Owing to the atomic level depth resolution of the technique, we show that T_C at the interface is the same as that in the inner layers, whereas, the temperature dependence of the magnetization is different. Moreover, it is revealed that the critical exponent exhibits a different behavior according to the easy axis of magnetization, which is in plane below $d_{\mathrm{Ni}} \sim 7$ MLs and is perpendicular to the surface above $d_{\mathrm{Ni}} \sim 8$ MLs.

II. EXPERIMENTS

All of our experiments were performed in an ultrahigh vacuum chamber with a base pressure of $\sim 1 \times 10^{-7}$ Pa, which was connected to the undulator beamline, BL-16A[17,18] of the Photon Factory at the Institute of Materials Structure Science, High Energy Accelerator Research Organization, Japan. A Cu(100) single crystal was cleaned by the repeated cycles of Ar^+ sputtering at 1.5 keV and subsequent annealing to ∼900 K. The Ni films were then grown on the Cu(100) substrate at room temperature by the electron bombardment evaporation from a Ni rod. The thickness of the Ni film was controlled by monitoring the oscillatory intensity of a reflection high-energy electron-diffraction spot. The films were subsequently covered with 3 MLs of Cu, which was evaporated by the electron bombardment heating of Cu wires in a Ta crucible.

The Cu/Ni/Cu(100) films were then investigated *in situ* by means of the depth-resolved XMCD technique by using the circularly polarized x rays from the APPLE-type undulators installed at BL-16A.[19] An experimental layout of the depth-resolved XMCD measurements is schematically illustrated in Fig. 1. In this technique, the probing depth of the XMCD spectrum is determined by the effective escape depth of the electrons λ_e, which depends on the electron emission

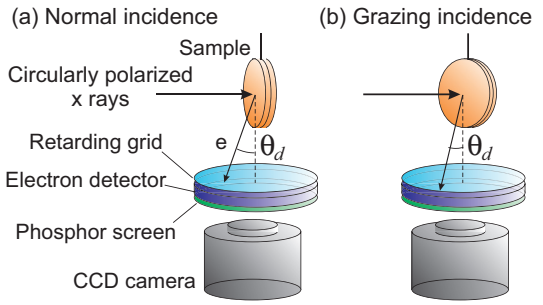

FIG. 1. (Color online) Schematic for the depth-resolved XMCD measurement in (a) normal and (b) grazing x-ray incidence configurations.

angle. The electrons emitted after x-ray absorption were separately collected at different detection angles θ_d by using an imaging-type detector, which consists of a microchannel plate, a phosphor screen, and a CCD camera. The partial electron yield mode with a retarding voltage of 500 V was adopted so that the Ni LMM Auger electrons were mainly collected. The probing depth λ_e was experimentally determined at each θ_d from the thickness dependence of the edge-jump intensity of the films.[11] One can obtain the depth-resolved XMCD spectra by analyzing a set of XMCD data recorded at different λ_e's.

The Cu/Ni/Cu(100) films are known to exhibit a spin reorientation transition from the in-plane to the perpendicular direction at ∼7-ML Ni thickness.[20] Therefore, normal incidence (NI) and grazing incidence (GI) x-ray configurations were adopted to investigate the perpendicularly (9-ML) and in-plane magnetized (5- and 6-ML) films, respectively, as illustrated in Fig. 1 because the XMCD signal reflects the magnetization component along the x-ray propagation direction. Note here that, since the probing depth is not controlled by the x-ray incidence angle but by the electron detection angle, this technique can be applied in both the NI and the GI configurations. The sample was mounted with $[1\bar{1}0]$ lying in the horizontal plane, and the angle between $[1\bar{1}0]$ and the x-ray beam was 30° at GI. A magnetic field of ∼500 Oe, which is always parallel to the incident x-ray beam, was applied by a yoke coil, and the coil was retracted out during the measurements. The temperature dependence was investigated both by increasing and by decreasing the sample temperature. We carefully checked that the same data were obtained even after heating the sample to ∼415 K, indicating that the Ni-Cu interdiffusion during the measurement is negligible in the present conditions. Although some intermixing between Ni and Cu can occur during the film growth process, it does not affect the relative temperature dependence, which is the main issue of the present study.

III. RESULTS AND DISCUSSION

A series of XMCD spectra, taken at different probing depths λ_e, ranging from ∼0.5 to ∼1.2 nm for the 9-ML Ni film is shown in Fig. 2. A large XMCD signal is observed at 298 K because the Curie temperature of the 9-ML film is estimated to be ∼410 K from the magnetization-temperature curve as shown later. The XMCD intensity slightly decreases as the probing depth decreases, which directly indicates that

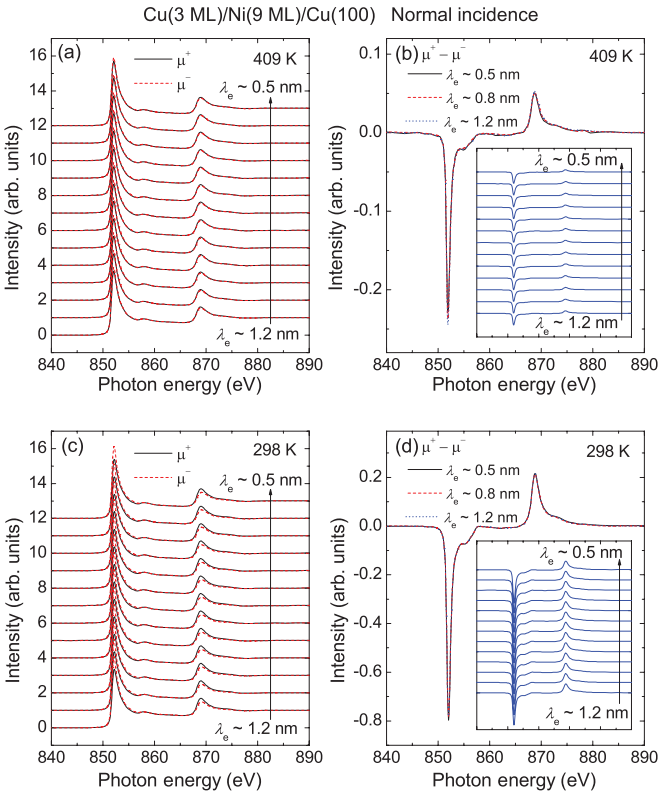

FIG. 2. (Color online) (a) Ni L-edge x-ray absorption spectra for the 9-ML Ni film taken with different probing depths, λ_e (corresponding to θ_d) recorded at (a) and (b) 409 and (c) and (d) 298 K. The x-ray absorption spectra with opposite circular polarizations are shown in (a) and (c), whereas, (b) and (d) correspond to their difference.

magnetization of the Ni layer at the interface to Cu is smaller than that in the inner Ni layers. Upon heating the film to 409 K, the XMCD signal significantly decreases. The XMCD intensity is smaller at the smaller probing depth, which is similar to the λ_e dependence at 298 K. However, here, it should be emphasized that, even at the smallest probing depth, the XMCD signal is clearly seen, although the sample temperature is close to the Curie temperature. This suggests that magnetization in the Ni layer at the interface to Cu still remains, even just below the Curie temperature of the film.

To further investigate the temperature dependence of magnetization, the XMCD spectra at the interface and in the inner layers are extracted from a set of data at different probing depths according to the following procedure.[13–16] The observed x-ray absorption spectrum $Y(E)$ is given by

$$Y(E) = C \sum_{i=1}^{m} \mu_i(E) \times \exp\left[-d\left\{\frac{i-1}{\lambda_e} + \frac{1}{\cos\theta}\sum_{k=1}^{i-1}\mu_k(E)\right\}\right], \quad (1)$$

where C, m, E, and θ are a normalization factor, the film thickness in ML units, the photon energy, and the x-ray incidence angle from the surface normal, respectively, and $\mu_i(E)$ is the absorption coefficient at the ith layer from the top Ni layer, which corresponds to the layer-resolved spectrum.

Then, $\mu_i(E)$'s are determined so as to reproduce the obtained set of spectra $Y(E)$ at different probing depths.

In the extraction process, the Ni film is divided into three regions, 1-ML-thick layers at the top and bottom interfaces to Cu and the remaining inner Ni layers. It is assumed that both interface layers have the same XMCD spectrum and that each layer in the inner layers shows the same spectrum, which is different from that at the interface. Unfortunately, such a model cannot include the effects of intermixing and/or roughness. However, it is always true that the extracted spectrum at the interface contains more contributions from the interface region, even if the interface is not sharp, than that from the inner layers. Therefore, the qualitative trend of the difference between the interface and the inner layers remains valid. Moreover, the degree of intermixing does not seem to change during the temperature-dependent measurements as mentioned above so that the relative temperature dependence is reliable. The adopted model is schematically illustrated in Fig. 3 together with the extracted XMCD spectra at different temperatures. It is clearly recognized that the XMCD signals both at the interface and in the inner layers almost simultaneously vanish between 409 and 415 K in the case of the 9-ML Ni film. This is also true for the 6-ML Ni film as shown in Fig. 3(b) in which the XMCD signals vanish at ~223 K. Thus, it is revealed that the interface layer has the same Curie temperature as that in the inner layers.

It is also recognized that the temperature dependences of the XMCD signal at the interface and in the inner layers are different from each other, especially for the 6-ML film.

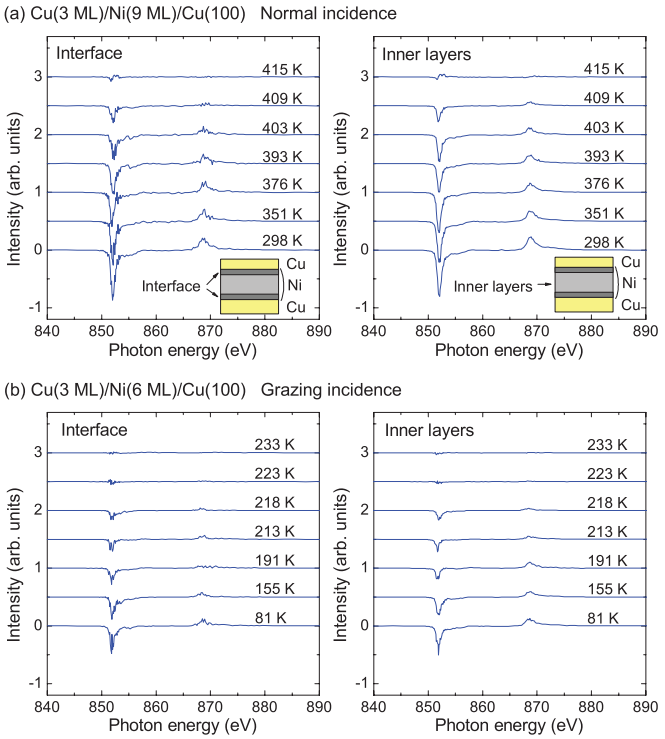

FIG. 3. (Color online) Temperature dependence of the interface and inner-layer components of the Ni L-edge XMCD spectra extracted from a set of XMCD data with different probing depths for (a) 9-ML and (b) 6-ML Ni films. The model used in the extraction procedure is schematically shown in (a).

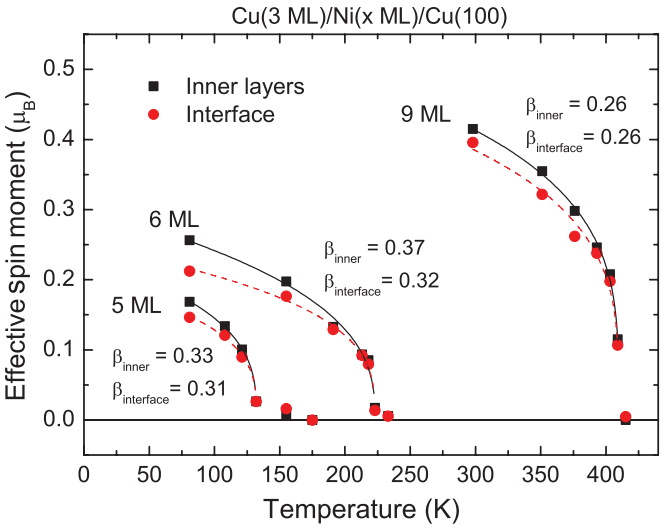

FIG. 4. (Color online) Effective spin moments in Ni at the interface and in the inner layers as functions of temperature, estimated from the sum-rule analysis for the extracted XMCD spectra. Each datum is fitted by Eq. (2), and the obtained critical exponents $\beta_{\text{interface}}$ and β_{inner} at the interface and in the inner layers, respectively, are indicated.

Figure 4 shows the temperature dependence of the effective spin moments at the interface and in the inner layers, estimated from the XMCD sum-rule analysis.[21,22] The obtained temperature dependence of spin moment $m(T)$ is fitted by using the following equation:[2]

$$m(T) = m_{\text{sat}}(1 - T/T_C)^{\beta}, \tag{2}$$

where m_{sat} represents the saturation spin moment, which corresponds to the moment at 0 K and β denotes the critical exponent. The estimated T_C is 132, 223, and 410 K for the 5-, 6-, and 9-ML Ni films, respectively.

First, let us discuss the in-plane magnetized 5- and 6-ML Ni films. As a basic trend, β decreases as the film thickness decreases, especially in the inner layers. This seems reasonable because the critical exponent is known to decrease with decreasing dimensionality. On the other hand, β at the interface is smaller than that in the inner layers, which might reflect the lower dimensionality at the interface compared with the inner layers. These trends can be more clearly recognized by plotting the normalized moment m/m_{sat} as a function of reduced temperature T/T_C as shown in Fig. 5. The smaller β at the interface is not consistent with a simulation, however, in which a simple mean-field theory is adopted as mentioned below. The temperature dependence of the remanent spin moment in the ith Ni layer is expressed as[4]

$$m(i,T) = \coth\left(\frac{H_i^{\text{eff}}}{k_B T}\right) - \frac{k_B T}{H_i^{\text{eff}}}, \tag{3}$$

where the effective field H_i^{eff} is defined as

$$H_i^{\text{eff}} = \sum_j J_{ij} m_j, \tag{4}$$

and J_{ij} represents the exchange-interaction constant between the Ni atom in the ith layer and the jth neighboring Ni atom with a remanent spin moment of m_j. By solving Eq. (3),

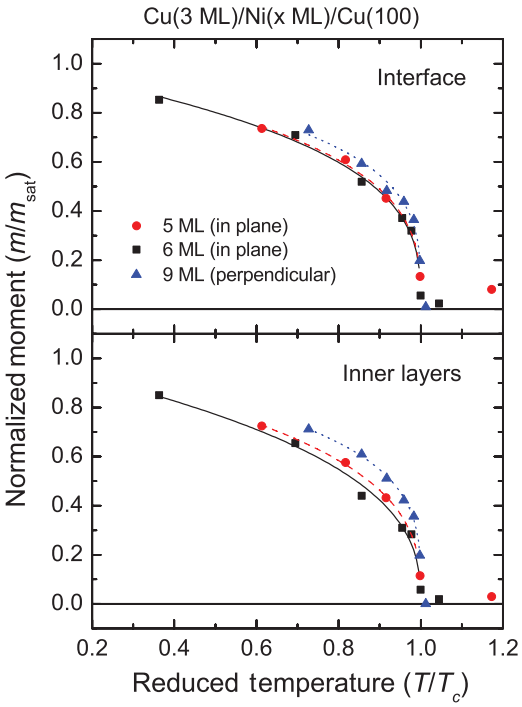

FIG. 5. (Color online) Temperature dependence of the effective spin moment at the interface (top) and in the inner layers (bottom) normalized by saturation moment m_{sat}, which is estimated by Eq. (2) as a function of reduced temperature T/T_C.

assuming the same J_{ij} value for all neighboring Ni atoms and considering only the nearest-neighbor atom pairs, we obtain $\beta_{\text{inner}} = 0.33$ and $\beta_{\text{interface}} = 0.42$ for 5 MLs, whereas, $\beta_{\text{inner}} = 0.37$ and $\beta_{\text{interface}} = 0.42$ for 6 MLs. Although the thickness dependence is consistent with our result, the difference between the interface and the inner layers shows an opposite trend from that obtained from the experiment. One of the simplest explanations for this discrepancy is that the exchange interaction constant J at the interface is larger than that in the inner layers. In fact, we obtain $\beta_{\text{inner}} = 0.20$ and $\beta_{\text{interface}} = 0.17$ for 5 MLs and $\beta_{\text{inner}} = 0.24$ and $\beta_{\text{interface}} = 0.20$ for 6 MLs if J in the interface layer is assumed to be twice that in the inner layers. No experimental or theoretical result has been reported, however, which revealed the enhancement of J at the Ni/Cu interface. On the contrary, it was reported that J, at the interface, is significantly smaller than that in the inner layers.[8] Some sophisticated theoretical treatment is necessary, which includes other effects, such as short-range spin ordering, layer-dependent magnetic anisotropy, and exchange interaction with farther atoms.

Let us turn onto the perpendicularly magnetized 9-ML Ni film. One can clearly find that β for the 9-ML Ni film is smaller than those for the 5- and 6-ML films. This seems strange because the dimensionality of 5- and 6-ML films is lower than that for the 9-ML film. This apparent discrepancy can be interpreted by the difference in the magnetization directions. In the in-plane magnetized films, the spin moment can exhibit at least four directions. On the other hand, the spin moment must be in either an up or a down state in the case of perpendicular magnetization, which can be regarded as the Ising model in which β is smaller than that in the Heisenberg model.[10] Thus, the increase in the degree of freedom would lead to the increase in the critical exponent of the in-plane magnetized films.

IV. SUMMARY

We have investigated, by means of the depth-resolved XMCD technique, the temperature dependence of the remanent magnetization of Ni thin films at the interface to Cu as well as that in the inner layers. Although magnetization at the interface is smaller than that in the inner layers, the Curie temperature, at which remanent magnetization vanishes, is revealed to be the same between the interface and the inner layers. In contrast, the critical exponent at the interface is slightly smaller than that in the inner layers, especially in the in-plane magnetized films. This difference cannot be explained by using a simple mean-field theory with the same exchange-coupling constant between the interface and the inner layers. It is also shown that the critical exponent for the in-plane magnetized film is significantly larger than that for the perpendicularly magnetized film due to the difference in the degree of freedom for the direction of the magnetic moment. The short-range spin ordering, layer-dependent magnetic anisotropy, and exchange interaction with farther atoms would be necessary to be taken into account in the comprehensive understanding of the observed behaviors.

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