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Anomalous magnetic phases in Fe/Cu(001) ultrathin films induced by CO adsorption

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Effects of CO adsorption on the magnetic properties of Fe films on Cu(001) were studied at 100 K as a function of the film thickness. CO adsorption does not affect the magnetic property of 2 ML Fe film but induces the spin reorientation from out-of-plane to in-plane magnetization for 3 and 4 ML films. It also causes the decrease of magnetization, which can be attributed to the reduction in surface magnetization. Fe films of 6 and 8 ML lose their magnetization by CO adsorption. Possible interpretations for these complicated magnetic behaviors are discussed.

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I. INTRODUCTION

Atomic and magnetic properties of Fe ultrathin films on Cu(001) have been extensively studied as a prototypical case of magnetic thin films.[1–16] Ultrathin Fe films (<11 ML) grown on Cu(001) exhibit an fcc phase even at room temperature[11] with an enlarged atomic volume of 12.1 Å^3, though the bulk Fe has an fcc phase in the temperature range of 1184–1665 K. These films have interesting magnetic and atomic structures depending on the thickness. In the film thickness less than 4 ML (regime I), the film exhibits a distorted fcc structure [face centered tetragonal (fct)] and a ferromagnetic coupling among a whole film with perpendicular magnetization. In the film thickness between 5 and 11 ML (regime II), the film shows fcc structure, and the top two layers couple ferromagnetically to each other and the inner layers are either antiferromagnetic below 200 K (the Néel temperature)[12] or in a spin-density wave state below 170 K.[13] In the film thickness above 11 ML (regime III), the phase transition occurs to a bcc state[14] and the magnetization easy axis changes to the in-plane direction. Recently, a bcc-like reconstruction at the surface was reported by scanning tunneling microscope observations in regimes I and II and such a reconstruction was suggested to be the origin of the ferromagnetic coupling.[10,15,16]

Effects of CO adsorption on the Fe films have been investigated by several researchers. Thomassen *et al.* reported that the Curie temperature of the film in regime II reduces upon CO adsorption.[11] Spisák and Hafner have performed first-principles calculations of clean and CO adsorbed Fe/Cu(001) films[17] and found that the bridge adsorption in molecular form is energetically preferred to hollow or on-top adsorption and that the magnetic moments of Fe films are substantially reduced. Tanabe *et al.* studied CO adsorption on Fe/Cu(001) films by infrared reflection-absorption spectroscopy. They found that CO molecules initially adsorb on bridge sites and then on on-top sites with increasing coverage.[18,19]

In this paper, we report the effects of CO adsorption on the magnetic properties of Fe/Cu(001) magnetic thin films as a function of film thickness. An Fe film of 2 ML is not affected by CO adsorption, while 3 and 4 ML Fe films change the magnetic properties drastically. The direction of magnetization rotates from perpendicular to in plane and the surface layer(s) loses the spin magnetic moment by CO adsorption. Consequently, only the two layers, which were located at the interface to the Cu(001) substrate, keep magnetization. Fe films of 6 and 8 ML lose ferromagnetic coupling of the top two layers by CO adsorption. Possible origins of these complex magnetic phases are discussed by considering the CO induced changes in the magnetic depth profiles and in low-energy electron diffraction (LEED) patterns, with the help of previous reports on the relation between the atomic structures and magnetic properties of the Fe films.

II. EXPERIMENT

All of our experiments were performed at BL-7A and BL-11A of the Photon Factory at the Institute of Materials Structure Science, High Energy Accelerator Research Organization, equipped with an ultrahigh vacuum chamber (a base pressure of 3×10^{-8} Pa).

Fe films were deposited at room temperature by an electron-beam evaporation on a Cu(001) single crystal, which was cleaned by repeated cycles of Ar$^+$ sputtering at 1.5 kV and annealing at 900 K. The film thickness was monitored with *in situ* reflection high energy electron diffraction (RHEED) observations and 2, 3, 4, 6, and 8 ML Fe films were prepared. The error in the thickness estimation is less than ~0.3 ML.

X-ray circular magnetic dichroism (XMCD) measurements were carried out at 100 K with synchrotron radiation emitted upward or downward from the electron orbit of the storage ring by 0.4 mrad, which is ~80% circularly polarized. The sample was magnetized using pulsed current through a coil (about 700 G) oriented along the x-ray propagation direction, and the measurements were performed at remanent conditions. Fe *L*-edge x-ray absorption spectra (XAS) were measured with the field antiparallel and parallel to the fixed photon helicity and XMCD spectra were ob-

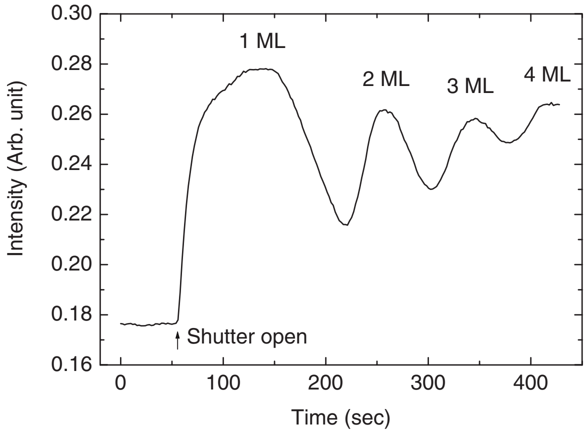

FIG. 1. A typical RHEED intensity curve observed *in situ* during Fe deposition on Cu(001).

tained from the difference of the two spectra. The direction of magnetization was examined using XMCD spectra at normal (90°) and grazing (30°) x-ray incidence, which are referred to as "NI" and "GI" hereafter. Depth-resolved XMCD measurements[20] were performed by an imaging-type microchannel plate detector with the partial electron yield mode, applying a retarding voltage of 500 V in order to collect Fe *LMM* Auger electrons. The probing depth was controlled by varying the electron detection angle. After each clean film measurement, the film was dosed with CO 5 L (1.3×10^{-5} Pa, 50 s) and the CO adsorbed films were studied in the same way as mentioned above.

LEED patterns were observed for clean and CO dosed Fe(2, 4 ML)/Cu(001) films.

III. RESULT AND DISCUSSION

A. Sample preparation

A typical RHEED intensity curve for the Fe growth is shown in Fig. 1. The ⟨100⟩ direction was aligned to the electron beam of RHEED. Good oscillations were observed for all the samples. Similar RHEED oscillations were reported in the previous study.[2] Thus, it is sure that layer-by-layer grown epitaxial Fe films were fabricated in our experiments.

C 1*s* x-ray photoelectron spectroscopy (XPS) spectra are shown in Fig. 2. The similar C 1*s* binding energies were reported in the previous studies for CO adsorption.[21,22] A peak at ∼285.5 eV corresponds to molecular CO, while a peak at ∼283 eV is attributed to atomic carbon. Since the XPS peak intensity for atomic carbon is less than a twentieth of that for molecular CO at saturation coverage (solid line), we can conclude that the effects of atomic carbon are negligible. In fact, the observed magnetic properties for the "clean" Fe film are consistent with the previous reports.[11,12,23] A similar result was obtained for O 1*s* XPS.

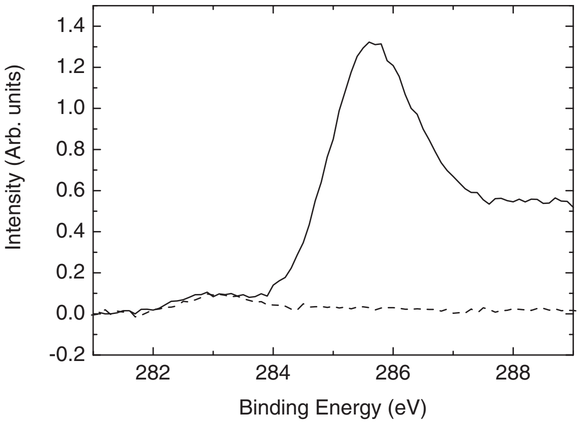

FIG. 2. The dashed line spectrum represents the C 1*s* XPS spectrum before CO exposure to Fe(2 ML)/Cu(001). The other solid line spectrum is the C 1*s* XPS spectrum of CO(5 L)/Fe(2 ML)/Cu(001).

B. Magnetic properties

Fe *L*-edge XAS and XMCD spectra of bare Fe(2 ML)/Cu(001) and Fe(4 ML)/Cu(001) are shown in Figs. 3(a), 3(b), 4(a), and 4(b), respectively. These spectra of 2 and 4 ML Fe films are quite similar to each other and the XMCD intensities for the GI spectra are weaker than those for the NI spectra by a factor of $\cos 60° = 1/2$, indicating that they have perpendicular magnetization. The spectra were analyzed to obtain a spin magnetic moment (m_s) by applying the sum rules,[24,25] leading to m_s=2.3 μ_B for Fe(2 ML)/Cu(001) and m_s=2.5 μ_B for Fe(4 ML)/Cu(001). The iron spin magnetic moment of Fe(3 ML)/Cu(001) was also obtained to be 2.4 μ_B (spectra not shown). Thus, all the clean films in regime I, which is in the thickness range up to 4 ML, have

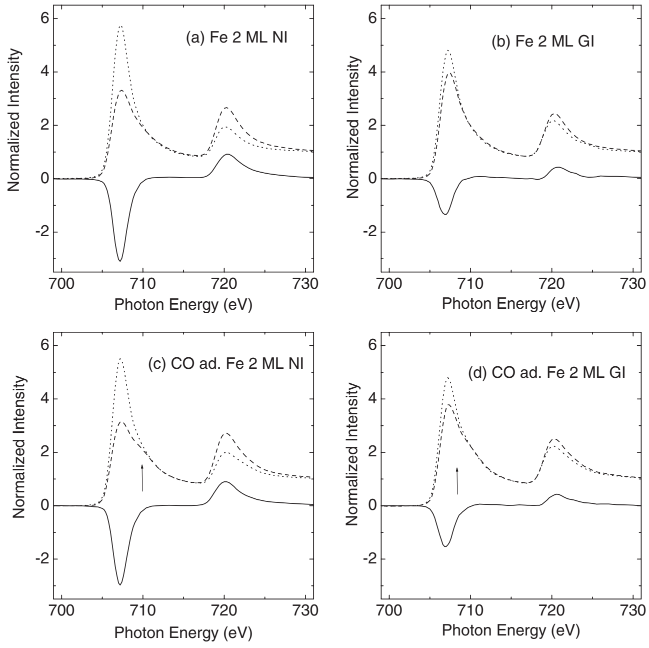

FIG. 3. Fe *L*-edge XAS and XMCD spectra of a 2 ML Fe film before and after CO adsorption: Fe(2 ML)/Cu(001) spectra in (a) NI and (b) GI geometries and CO/Fe(2 ML)/Cu(001) spectra in (c) NI and (d) GI geometries. The solid lines represent XMCD difference spectra, while the dashed and dotted lines correspond to parallel and antiparallel configurations between the photon helicy and majority spin. Arrows in (c) and (d) indicate a shoulder structure at 710 eV.

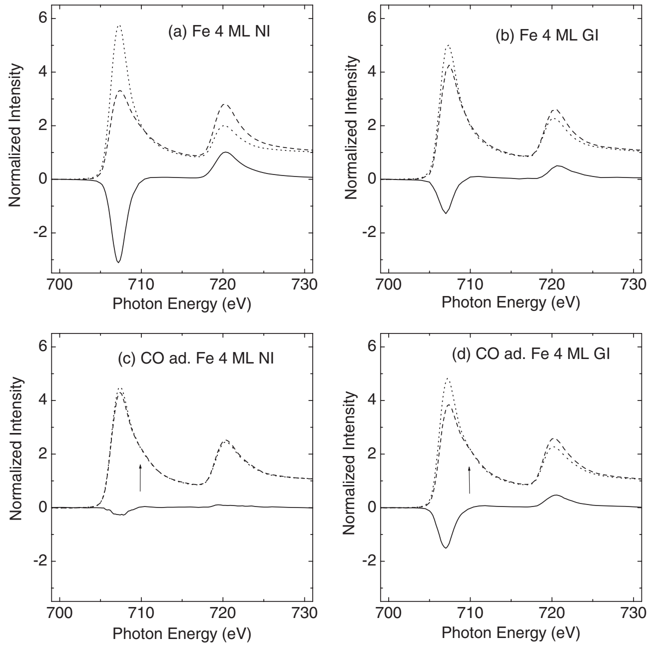

FIG. 4. Fe *L*-edge XAS and XMCD spectra of a 4 ML Fe film before and after CO adsorption: Fe(4 ML)/Cu(001) spectra in (a) NI and (b) GI geometries, and CO/Fe(4 ML)/Cu(001) spectra in (c) NI and (d) GI geometries. The solid lines represent XMCD difference spectra, while the dashed and dotted lines correspond to parallel and antiparallel configurations between the photon helicy and majority spin. Arrows in (c) and (d) indicate a shoulder structure at 710 eV.

perpendicular magnetization and the spin magnetic moment of about 2.4 μ_B. These properties indicate a ferromagnetic coupling among the whole layers and are in good agreement with the previous experiment.[11]

Next, let us discuss the effect of CO adsorption on the Fe films. Fe *L*-edge XAS and XMCD spectra of CO/Fe(2 ML)/Cu(001) are shown in Figs. 3(c) and 3(d), and those of CO/Fe(4 ML)/Cu(001) in Figs. 4(c) and 4(d). These XAS spectra have shoulder structures at 710 eV, as indicated by arrows. This shoulder could originate from CO adsorbed Fe atoms because this is not observed in the clean film spectra shown in Figs. 3(a), 3(b), 4(a), and 4(b).

The XMCD spectra of CO/Fe(2 ML)/Cu(001) are substantially the same as those of Fe(2 ML)/Cu(001): the same intensities and magnetization direction. CO adsorption does not affect the XMCD spectra of the 2 ML Fe film. In contrast, the XMCD spectra from CO/Fe(4 ML)/Cu(001) exhibit almost no signal for the NI geometry but significant signal for the GI geometry. This result directly indicates that the film has in-plane magnetization. Thus, the CO adsorption rotates the easy axis of magnetization of the 4 ML film from perpendicular to in plane. Note that the XMCD signal intensity of CO/Fe(4 ML)/Cu(001) is much smaller than that of CO/Fe(2 ML)/Cu(001). The XMCD spectra were analyzed in the same way as mentioned above, and we obtained $m_s = 2.3\ \mu_B$ for CO/Fe(2 ML)/Cu(001) and $m_s = 1.1\ \mu_B$ for CO/Fe(4 ML)/Cu(001). The spin magnetic moment of CO/Fe(4 ML)/Cu(001) is about half of that of CO/Fe(2 ML)/Cu(001). Although the spectra are not shown, the spin magnetic moment of CO/Fe(3 ML)/Cu(001) was also obtained to be 1.5 μ_B.

TABLE I. Layer-averaged spin magnetic moments of Fe in Fe/Cu(001) and CO/Fe/Cu(001) films. "⊥" and "∥" represent perpendicular and in-plane magnetizations, respectively. "SRT" is an abbreviation of spin reorientation transition.

Fe regime	Fe (ML)	Clean Fe	CO 5 L dosed Fe	
Regime I	2	2.3 μ_B, ⊥	2.3 μ_B, ⊥	
	3	2.4 μ_B, ⊥	1.5 μ_B, ∥	SRT
	4	2.5 μ_B, ⊥	1.1 μ_B, ∥	SRT
Regime II	6	⊥	No mag.	
	8	⊥	No mag.	

For 6 and 8 ML Fe films (regime II), the films exhibited perpendicular magnetization and ferromagnetic coupling between the surface two layers (spectra not shown), as reported in the previous papers.[12,23] CO adsorption on these films resulted in the disappearance of the XMCD signal. This disappearance of XMCD could indicate no ferromagnetic coupling at 100 K. In fact, it was reported that 0.5 L CO exposure lowers the Curie temperature of the Fe film in regime II by 40 K.[11] We cannot exclude, however, a possibility that the magnetic field of 700 G was not enough to obtain the XMCD signal, due to the increase in the coercive field by CO adsorption.

The obtained magnetic properties of all the films are listed in Table I with the magnetization directions for clean and CO adsorbed films. These spin magnetic moments are layer-averaged values. Note that the CO adsorption decreases the spin moment more significantly for the thicker Fe film. No essential change occurs for the 2 ML film, but the averaged spin moment reduces from 2.4 μ_B to 1.5 μ_B for the 3 ML film and from 2.5 μ_B to 1.1 μ_B for the 4 ML film, and for the 6 and 8 ML films, the magnetization disappears. These results seem rather strange because the Curie temperature of thin films should increase as the thickness increases.

Here, we discuss three phenomena resulted from CO adsorption: (1) the apparent reduction of spin magnetic moment at 3 and 4 ML, (2) the reorientation of the magnetization direction observed also in 3 and 4 ML films, and (3) the disappearance of ferromagnetic coupling in 6 and 8 ML films.

Let us start with a hypothesis that CO adsorption would reduce ferromagnetic coupling of the surface two layers since no magnetization was observed in CO/Fe(6 and 8 ML)/Cu(001) films. This hypothesis can explain the apparent reduction of the spin magnetic moment observed in CO/Fe(4 ML)/Cu(001); the surface and subsurface layers lose their magnetization by CO adsorption, and only the bottom two layers keep magnetization. In fact, the apparent spin magnetic moment of 1.1 μ_B is about half of the moment (2.5 μ_B) observed before CO adsorption.

Another hypothesis is required to understand the results for CO/Fe(2 ML)/Cu(001) because the film has a large magnetic moment even after CO adsorption. Then, we as-

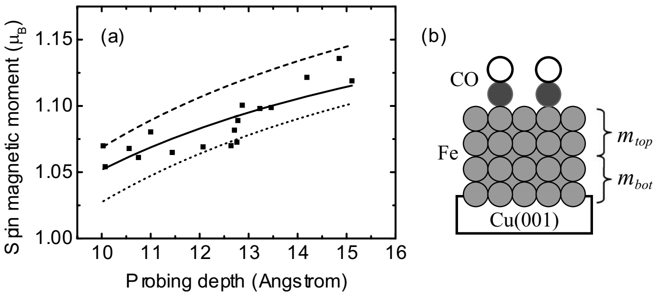

FIG. 5. (a) The probing depth dependence of the Fe spin magnetic moment m_s of CO/Fe(4 ML)/Cu(001). Filled squares are observed data, while the solid line is a fitting curve with $(m_{\text{top}}, m_{\text{bot}}) = (0.17, 2.31)\mu_B$. Dashed and dotted lines are simulated curves with $(m_{\text{top}}, m_{\text{bot}}) = (0.0, 2.6)\mu_B$ and $(0.0, 2.5)\mu_B$, respectively. (b) A schematic model for the simulations and fittings.

sume that the magnetic structure of two layers adjacent to the Cu(001) substrate is kept by a certain Cu(001) substrate effect. This is to say that the Cu(001) substrate has a stronger effect on the magnetization of the nearest and next nearest two Fe layers than the CO adsorption. Such an effect might come from the in-plane lattice expansion related to the epitaxial growth of Fe films.

This hypothesis can also explain the other case of CO/Fe(3 ML)/Cu(001). The surface two layers tend to lose their magnetization but the Cu(001) substrate keeps the magnetization of the bottom two layers. Consequently, only the surface single layer loses magnetization and the rest two layers exhibit magnetization even after CO adsorption. The obtained moment of $1.5\mu_B$ agrees with the expected value of the spin magnetic moment, $1.6\mu_B$, which is two-thirds of $2.4\mu_B$.

These two hypotheses were proved by using the depth-resolved XMCD technique,[20] which makes use of the probing depth dependence of the XMCD spectra on the Auger electron emission angle. The probing depth dependence of the obtained spin magnetic moment of CO adsorbed 4 ML Fe is shown in Fig. 5(a). The spin magnetic moment decreases with decreasing probing depth, directly showing that the moment is small near the surface.

We analyzed the depth-resolved XMCD data from CO/Fe(4 ML)/Cu(001) with a simple model, which is composed of the top two layers and bottom two layers, as shown in Fig. 5(b). In this model, the observed spin magnetic moment, $m(x)$, at a probing depth x is expressed by m_{top} and m_{bot}, which represent the spin magnetic moments of the top two layers and bottom two layers, respectively. The interlayer distance was fixed to 1.8 Å. Simulated results are also shown in Fig. 5(a), which clearly show that the magnetization of CO/Fe(4 ML)/Cu(001) locates in the bottom two layers, while the top two layers are almost nonmagnetic. We cannot exclude a possibility that the top two layers couple antiferromagnetically, but in any case, the sum of the magnetization of the top two layers should be small.

C. Low-energy electron diffraction patterns

LEED patterns of clean and CO dosed Fe(2, 4 ML)/Cu(001) films were shown in Fig. 6, where weak but apparent (4×1) or (5×1) patterns are recognized. These patterns agree with the previous studies,[7,9,10,26] in which (4×1), (5×1), and (6×1) LEED patterns were reported depending on the thickness and growth condition. In the STM study,[10] the $(n\times1)$ patterns were related to a bcc-like reconstruction at the film surface. It was suggested that the bcc-like structure is the origin of the ferromagnetic coupling in the films.

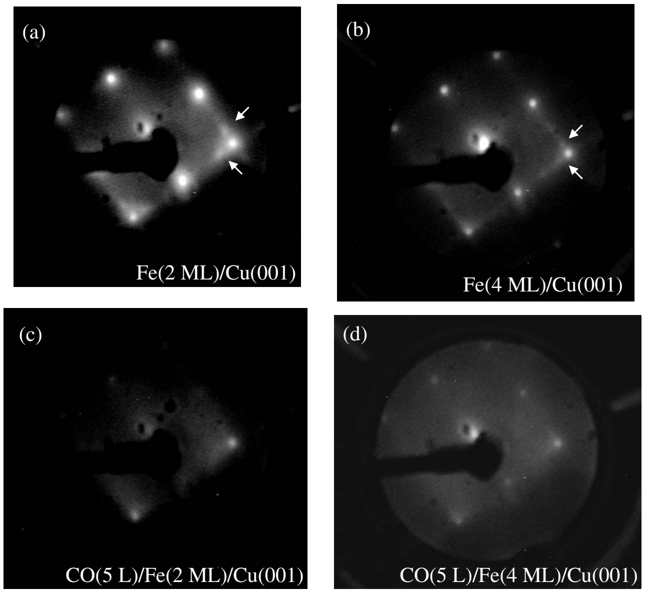

FIG. 6. LEED patterns of (a) Fe(2 ML)/Cu(001), (b) Fe(4 ML)/Cu(001), (c) CO/Fe(2 ML)/Cu(001), and (d) CO/Fe(4 ML)/Cu(001). Electron energy was 160 eV. White indicates extra spots, such as (4×1) and (5×1).

For the CO covered films, on the other hand, it is difficult to find (4×1) or (5×1) patterns because of very defused and weakened spots. The LEED patterns of the CO/Fe(2 ML)/Cu(001) film, however, exhibit a little bit visible spots or streaks than that of the CO/Fe(4 ML)/Cu(001) film. This result suggests that the atomic structure of the CO/Fe(2 ML)/Cu(001) film is not seriously deviated from that of clean 2 ML Fe, while the 4 ML Fe film is probably affected by adsorbed CO to change its atomic structure. In fact, the 4 ML Fe film was reported to be more unstable than thinner films.[9] Note here that no extra spots such as (2×1) or $c(2\times2)$ can be found. This is in clear contrast to the previous report,[2] where Fe films were grown under CO exposure and a $c(2\times2)$ superstructure was observed. Moreover, since a (2×1) pattern was reported for the Fe films in regime II,[26] a CO induced phase transition to regime II can be excluded. Therefore, one can consider that the remained magnetization of 2 ML Fe film after CO adsorption is attributed to the relatively unchanged atomic structure of the film and that the reduced magnetizations of 3 and 4 ML films are possibly due to some reconstruction of their atomic structure.

D. Suggestions for the relation between the atomic structures and spin reorientation transition

Let us discuss the possible origin of the CO induced changes in the magnetic properties, especially the spin reorientation transition (SRT), observed in the 3 and 4 ML films. The magnetization anisotropy energy (MAE) of a thin film per unit volume is usually described as

$$\Delta E = -2\pi M_s^2 + (K_i + K_s)/d + K_v, \tag{1}$$

where K_v, K_i, and K_s are volume, interface, and surface contributions to the second order MAE K_2, respectively, and M_s is the magnetization. The origin of the adsorbate-induced SRT has been discussed in terms of the atomic structure change[28] or the reduction of surface magnetization.[29,30] In the latter case, only the surface MAE K_s, is essentially affected, while all of the MAE terms including K_v can be changed in the former case. In fact, the H adsorption-induced SRT of Ni/Cu(001) films was related to the adsorbate-induced layer relaxation of tetragonally distorted Ni films.[28]

For the Fe/Cu(001) films, Platow *et al.* reported that $K_v = 77.7\ \mu\text{eV/atom}$ and $(K_i+K_s)/2 = 120\ \mu\text{eV/atom}$.[27] Note here that the "surface" anisotropy, K_2^s in their notation, is an average between the surface and interface components, $(K_i + K_s)/2$. Although the $(K_i+K_s)/2$ exhibits a strong tendency to perpendicular magnetization, one cannot determine which of the surface and interface mainly contributes perpendicular magnetic anisotropy (PMA).

First, we consider a simple case, where the surface magnetization of the Fe films is reduced upon CO adsorption, keeping the atomic structure and magnetic property of the inner layers unaffected. Let us assume that the Fe surface has a large PMA and this is the key to perpendicular magnetization of Fe(2–4 ML)/Cu(001) films. In fact, a MAE of 174 μeV/atom was reported for the surface layer of Fe(2 ML)/Ni/Cu(001),[31] though the atomic structure and MAE might be different from the present sample, Fe/Cu(001). In this case, CO/Fe(4 ML)/Cu(001) should lose the PMA originating from K_s because the top 2 ML has no magnetization after CO adsorption, and this film would undergo SRT to in-plane magnetization. This is a possible way to explain the SRT we observed in the present study.

Next, we discuss possible atomic structure changes upon CO adsorption. Unfortunately, we cannot provide quantitative information on the crystalline structure in the present study, but we could try to explain the origins of spin reorientation transition based on relevant literatures. Shen *et al.* reported the in-plane anisotropy of fcc Fe/Cu(001) films grown by pulsed laser deposition (PLD) method, in the thickness range of 2–5 ML.[32] They claimed that PLD films have an almost perfect fcc structure, while thermal deposition films are the mixed structure of fct and fcc. Another report[33] by Pan *et al.* revealed that Fe films on Fe_xMn_{1-x}(17 ML)/Cu(001) show various structures and magnetic properties depending on x. Fe films on Fe_xMn_{1-x}(17 ML)/Cu(001) with $x \leqq 0.35$ have the bcc structure and in-plane magnetization, while Fe films on Fe_xMn_{1-x}(17 ML)/Cu(001) with $x \geqq 0.35$ show the fcc structure and are not ferromagnetic.

Therefore, if the bottom part of our CO/Fe(3,4 ML)/Cu(001) films has the perfect fcc structure like the PLD films, the film can exhibit in-plane magnetization. Although it is possible to consider that the bottom part is in an in-plane magnetized bcc phase as in Fe/FeMn/Cu(001), it seems unlikely because our LEED patterns show any evidence for the bcc structure. For the top part of the CO/Fe(3,4 ML)/Cu(001) films, in which a significant reduction in magnetization was observed as described above, it may be a possible conclusion that the top part is in a nonferromagnetic imperfect fcc phase. In fact, a first-principles local-spin-density investigation of clean and CO adsorbed 4 ML Fe films on Cu(001) (Ref. 17) showed a large reduction of the magnetic moment not only for the top layer, but a larger decrease is reported in the second layer. The decrease in the magnetic moments is accompanied by a reduction of the interlayer spacings.

E. Comparison with the CO-assisted growth

Finally, we compare the present results with the previous studies,[2,11] which reported the magnetic and atomic structures of Fe films grown under CO exposure. Thomassen *et al.* showed that the layer-by-layer growth accompanied with perpendicular magnetization, which is characteristic of regime II, is expanded to ~13 ML. They also reported, however, that such an expansion of regime II is not induced by CO adsorption after the film deposition. Kirilyuk *et al.* showed that the layer-by-layer growth is continued to even 30–35 ML, but they also suggested a dissociation of CO molecule. In contrast, molecular CO adsorption was confirmed by x-ray photoemission and x-ray absorption spectroscopies in the present study, in which CO was exposed at ~100 K after the film deposition. Therefore, it is inappropriate to directly compare the present results to those obtained by CO exposure during the film growth because of the different film structure and chemical species at the surface.

Nevertheless, it is interesting that in-plane magnetization in 2–5 ML Fe films was observed for the C_2H_2-assisted growth.[2] The origin of the SRT, however, seems different from the present case, in which surface magnetization is almost completely killed by CO adsorption, since no significant reduction in the magnetic moment was reported there.

IV. SUMMARY

In summary, the effects of CO adsorption on the magnetic properties of Fe(2, 3, 4, 6, and 8 ML)/Cu(001) films were studied at 100 K. The 2 ML film is not affected by CO adsorption. In contrast, the 3 and 4 ML Fe films change their magnetic properties drastically. The direction of magnetization rotates from perpendicular to in plane. In addition, the surface layer(s) loses the spin magnetic moment upon CO adsorption and consequently only the bottom two layers, which are located at the interface to the Cu(001) substrate, have magnetization. Magnetization of 6 and 8 ML Fe films disappears after CO adsorption, in which only the top two layers have magnetization before CO adsorption. These complex magnetic phases can be interpreted by assuming that CO adsorption kills ferromagnetic coupling of surface layer(s), while the Cu(001) substrate keeps the magnetic structure of adjacent two layers. The LEED results suggest that the atomic structure of the 4 ML Fe film is changed by CO adsorption, while that of the 2 ML film is not seriously affected.

The rotation of the magnetization direction observed in the CO/Fe(3,4 ML)/Cu(001) films can be attributed to the decrease in surface PMA due to the surface demagnetization and/or some structure changes. With the help of relevant literatures, it might be a possible interpretation that the top part of the CO adsorbed film loses its magnetization accompanied with a reduction of interlayer spacing, while the bottom part has an almost perfect fcc structure with in-plane magnetization. We should emphasize here that the present phenomena cannot be explained by an expansion of regime II to thinner region such as 3 or 4 ML. In regime II, the ferromagnetic coupling exists near the surface, while the present results for CO/Fe(4 ML)/Cu(001) show that the surface layers are magnetically dead. Moreover, the SRT to in-plane magnetization cannot be explained by the phase transition to regime II.

Thus, the various magnetic phases observed in our experiment might have different crystalline structures, though we could not exactly determine the film structures. The next stage of this discussion requires very precise information on the crystalline structures of CO adsorbed Fe films.

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