Extremely sharp spin reorientation transition in ultrathin Ni films grown on Cu(110)-(2×1)O

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We report a combined reflectance difference spectroscopy and scanning tunneling microscopy study revealing the extreme sensitivity of the magnetic properties of Ni films on Cu(110)-(2×1)O near the spin reorientation transition. Upon completion of the ninth contiguous Ni monolayer (ML) the magnetic easy axis switches from in-plane to out-of-plane within a narrow thickness range of at most 0.1 ML. In addition, we find that the reversible adsorption of CO completely quenches the out-of-plane magnetization of these Ni films. As little as 0.1 L of CO is sufficient for spin reorientation at a film thickness of 9.1 ML.

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During the last decade ultrathin ferromagnetic films on nonmagnetic substrates have been an active area of research. The spin reorientation transition (SRT), i.e., the spontaneous change in the magnetization direction, in particular from in-plane to out-of-plane, is one of the most interesting effects occurring in such thin films.[1,2] The SRT can be driven by various parameters such as the film thickness,[1,3] temperature,[1] stress,[4] or adsorbate coverage.[5,6]

As a model system, Ni grown on Cu(001) has been thoroughly investigated due to its favorable growth mode resulting in rather smooth magnetic films. For instance, a thickness driven SRT around 8–11 ML (Ref. 7) and an adsorbate (H_2, CO) induced SRT from in-plane to out-of-plane magnetization[5,7] have been reported. When Ni is deposited on an oxygen precovered Cu(001) surface, the oxygen acts as a surfactant, leading to an improved layer-by-layer like film growth. As a result, the SRT is shifted to about 5 ML lower Ni coverages, which clearly demonstrates the strong interplay between film morphology and magnetism.[8,9]

On the other hand, Ni films grown on the pristine Cu(110) surface have received much less attention due to the less favorable 3D-type growth mode. Earlier studies provided contradicting results about the existence of an out-of-plane magnetic regime.[10,11] More recently, it has been shown that for Ni grown on bare Cu(110) the magnetic easy axis always lies in the surface plane.[12] The out-of-plane magnetic phase found by Wu *et al.*[10] was attributed to oxygen contamination, since on Cu(110), like on Cu(100), oxygen acts as a surfactant for the Ni growth. This fact can be exploited by using the oxygen precovered Cu(110)-(2×1)O surface as a template. After an initial rough phase the growth mode changes at 6–8 ML (Refs. 13 and 14) to a layer-by-layer like mode where the oxygen floats on top and induces a (2×1) reconstruction of the topmost Ni layer. The so grown Ni films undergo a SRT between 7–7.5 ML to an out-of-plane magnetic phase. Above 35 ML, the magnetic easy axis slowly tilts back into the surface plane.[12,15]

Although the influence of the film morphology on the magnetic properties of the Ni/Cu(110)-(2×1)O system can thus be conjectured, the actual origin of the SRT is still not understood. In particular, it is not clear whether the SRT at 7–7.5 ML (Refs. 12 and 15) is directly related to the smoothing of the film, which should also occur around 6–8 ML.[13,14] (Note that, for consistency with the usual definition of 1 ML, the above values and all thicknesses reported in the earlier works[12–15] should be multiplied by $\sqrt{2}$, not by a factor of 2 as stated in Ref. 12.)

In the present study, we have combined scanning tunneling microscopy (STM) and reflectance difference–magneto-optical Kerr effect (RD-MOKE) spectroscopy to accurately determine the correlation between magnetism and morphology near the SRT for the Ni/Cu(110)-(2×1)O system. We will show that the Ni film is already smooth before the onset of the SRT, which takes place at a critical thickness of 9 ML and which turns out to be much sharper than presumed earlier. Furthermore, we have investigated the effect of CO adsorption on the magnetic properties of the Ni films. We find that CO adsorption induces a fully reversible SRT back to an in-plane magnetic phase. The tiny amounts of CO which suffice to obtain a complete spin reversal just above the critical thickness further corroborate the extreme sharpness of the SRT.

The experiments presented below were carried out in a UHV chamber with a base pressure below 1×10^{-10} mbar and equipped with a variety of surface analytic tools, namely, a low-energy electron diffraction (LEED) and Auger electron spectroscopy (AES) system, an ion gun and gas inlet for sample preparation, a quadrupole mass analyzer, a quartz crystal microbalance, and a homebuilt variable temperature STM operating at 12–500 K. The setup further includes a metal deposition source (Focus EFM3T triple evaporator), a strain free quartz window for optical access perpendicular to the sample surface, and a homebuilt *in situ* electromagnet. The single-coil design allows concurrent optical probing and metal deposition on the sample, while applying magnetic fields up to ±40 mT perpendicular to the sample surface. The high-quality single-crystal Cu(110) sample (miscut angle $<0.1°$) is mounted on a manipulator where its temperature can be varied between 12 and 1000 K by means of a continuous flow liquid He cryostat and electron impact heating. The sample holder itself is built from copper to avoid ferromagnetic materials near the sample. The STM investigations were performed on the same sample holder/manipulator combination. Thus, the present system allows magneto-optical and STM characterization on the same sample without any transfer and under permanent control of the experimental parameters.

In the present study we have employed reflectance difference spectroscopy (RDS/RAS), an optical technique, which

measures the normalized difference of the reflectivity at normal incidence for light polarized along two mutually perpendicular orientations (x,y):

$$\frac{\Delta r}{r} = 2\frac{r_x - r_y}{r_x + r_y}. \quad (1)$$

In the present case x and y are aligned with the major in-plane crystallographic axes of the sample, i.e., the $[1\bar{1}0]$ and the [001] directions, respectively. RDS has proven to be a versatile tool to investigate various processes on anisotropic surfaces such as surface reconstructions, adsorption, or thin-film growth.[16–19] If the sample is magnetic, RDS can be used to simultaneously probe its magnetic properties via the polar magneto-optical Kerr effect (MOKE).[12,15] Despite its limitations to out-of-plane magnetization, RD-MOKE is an extremely powerful and convenient technique. The setup used in the present work spans an optical range from 1.5 to 5.5 eV and simultaneously measures the real and imaginary parts of the RD signal $\Delta r/r$, thus providing access to both Kerr rotation α and Kerr ellipticity ϵ over an extended spectral range. The magnetic contribution can be separated from the conventional RD signal by applying an external magnetic field to the sample which switches the magnetization direction between pointing into $(\downarrow)$ and out of $(\uparrow)$ the surface plane, respectively:

$$\alpha + i\epsilon = \frac{1}{4}\left[\frac{\Delta r}{r}(\uparrow) - \frac{\Delta r}{r}(\downarrow)\right]. \quad (2)$$

The Cu(110) surface was prepared by 900 eV Ar ion sputtering and subsequent annealing to 800 K. After sample preparation no impurities could be detected by AES and LEED images revealed a sharp (1×1) diffraction pattern. The clean surface was then exposed to about 40 L of molecular oxygen. After subsequent annealing to 600 K, LEED revealed a sharp (2×1) pattern characteristic of a well-ordered Cu(110)-(2×1)O surface. Ni was deposited via e-beam evaporation from a high purity Ni rod. All Ni films investigated here were grown at 300 K at a rate of 0.5 ML/min which was stabilized by regulating the ion flux. Thickness calibration was achieved by correlating RD transients and STM images recorded at different coverages with the deposition rates determined before and after deposition by means of a calibrated quartz microbalance.

Figure 1(a) shows a set of characteristic RD transients recorded at fixed photon energy of 3.5 eV as a function of Ni coverage. The three curves show the evolution of the RD signal in zero field as well as in the presence of a tiny magnetic field of 0.3 mT (3 Oe) applied during deposition along the $\downarrow$ and $\uparrow$ direction, respectively. The photon energy of 3.5 eV was chosen to obtain the strongest magnetic response. Up to 9 ML the three transients perfectly overlap, indicating that no out-of-plane magnetization is present. At 9 ML the three curves split due to the sudden onset of the out-of-plane magnetization. While the transient for zero field varies smoothly and continues to represent the morphology related RD contribution only, the lower and the upper branches carry an additional magnetic (MOKE) signal which starts abruptly

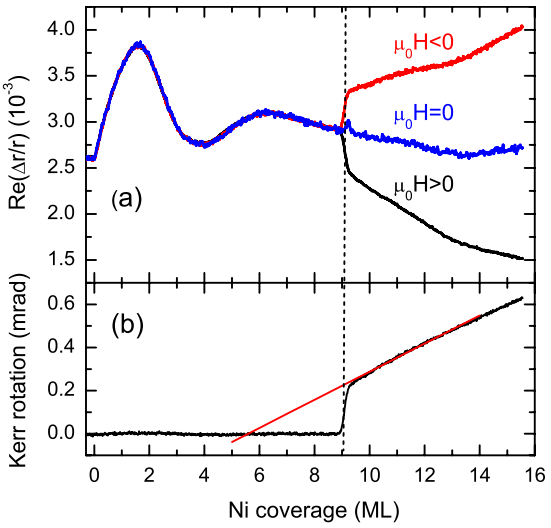

FIG. 1. (Color online) (a) RD transients recorded at 3.5 eV during the deposition of Ni on the Cu(110)-(2×1)O surface at 300 K with different magnetic fields applied: $\mu_0 H \approx +0.3$ mT (black), $\mu_0 H \approx -0.3$ mT (red), and $\mu_0 H = 0$ mT (blue). (b) Purely magnetic (MOKE) contribution to the RD signal as a function of film thickness. The linear extrapolation (solid line) suggests the presence of five to six nonmagnetic layers.

when the SRT is reached at 9 ML. The sign of the magnetic contribution is either positive or negative depending on the orientation of the external magnetic field. STM images confirmed that the SRT always occurs at the same Ni coverage making the "jump" in the RDS signal an extremely precise $(<1\%)$ measure of the critical thickness Θ_c for the onset of the SRT.

Postgrowth hysteresis measurements show that the upper and lower transients correspond to a fully magnetized Ni film. From the difference of these "magnetic" transients we thus obtain the saturation value of the remanent Kerr rotation [Eq. (2)] once the easy magnetization axis switches to out-of-plane at the critical film thickness Θ_c. The variation in the remanent magnetization as a function of the film thickness is shown in Fig. 1(b). While from 0–9 ML no remanent out-of-plane magnetization exists, the Kerr rotation above 9.25 ML increases linearly as expected in the thin-film limit. Extrapolation to zero remanence suggests the presence of five to six nonmagnetic layers as reported previously by Herrmann *et al.*[12] The SRT occurs in an extremely narrow range between 9 to 9.25 ML and the Kerr rotation increases almost spontaneously. In fact, 0.25 ML is an upper limit for the coverage range over which the SRT occurs, since it could be broadened, e.g., as a consequence of a slightly inhomogeneous evaporation rate across the spot size of the RDS beam which was about 6–7 mm in this experiment. We estimated the effect of a laterally inhomogeneous evaporation by scanning the magnetic properties across the sample using a reduced RDS spot diameter of about 1.5–2 mm. From the linear relationship of the magnetization above the SRT [Fig. 1(b)] we could estimate the maximum coverage variation across the sample to be below 0.2 ML for a 9 ML thick film, which is about the same value we found for the width of the SRT measured with the larger spot size. Moreover, no local coex-

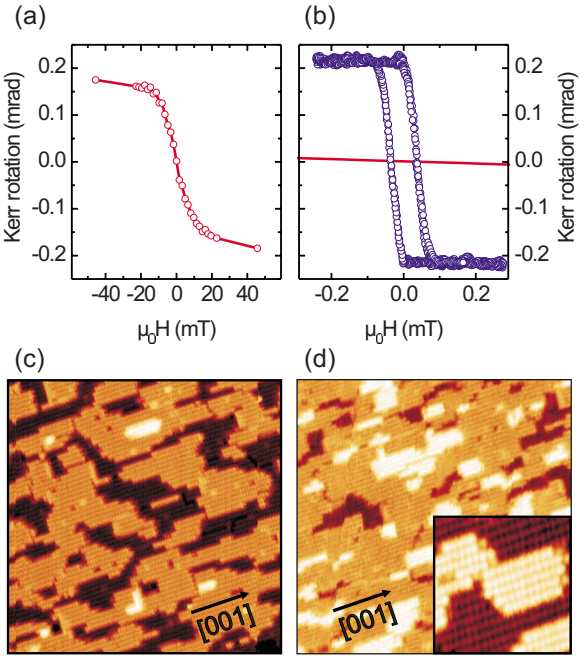

FIG. 2. (Color online) Hysteresis loops for (a) 8.65 ML and (b) 9.08 ML of Ni on Cu(110)-(2×1)O. To illustrate the dramatic change in the magnetic properties across the SRT, the hysteresis loop for 8.65 ML (a) is also shown in (b) as a solid line for comparison. (c),(d) Corresponding STM images of the 8.65 ML and the 9.08 ML films, respectively (scan size: 30×30 nm^2). The inset in (d) shows a 7×7 nm^2 detail of a 9 ML thick film with atomic resolution revealing the (2×1)O reconstruction of the topmost Ni layer.

istence of in-plane and out-of-plane magnetization could be detected when using the small RDS spot size. This suggests that the actual coverage range over which the SRT occurs locally should be less than the detection limit of about 0.1 ML.

Additional experiments were also conducted after stopping the deposition in the vicinity of the SRT and watching the temporal evolution of the magnetic RD signal. In this case the onset of the critical coverage is reduced by 0.15 ML to 8.85 ML and pure out-of-plane magnetism is observed at 9.08 ML as shown in the hysteresis loop of Fig. 2(b). Below 8.85 ML we observe hard axis loops [Fig. 2(a)]. In the narrow range from 8.85 to 9 ML the out-of-plane magnetization develops within 2–4 min. We attribute this to the smoothing kinetics of the Ni films at room temperature.

STM images of Ni films with coverages just below and above the SRT are shown in Figs. 2(c) and 2(d), respectively. The presence of only three uncovered layers in both images clearly demonstrates that the change to a layer-by-layer like growth and the concomitant smoothing of the Ni film is already accomplished before the onset of the SRT. Therefore, the observed SRT is purely thickness driven and not induced by additional morphological changes. On the basis of the results presented in Figs. 1 and 2, the occurrence and the sharpness of the SRT in the Ni/Cu(110)-(2×1)O system can now be linked unambiguously to the formation of a contiguous ninth monolayer of Ni.

A further demonstration of the extremely high sensibility

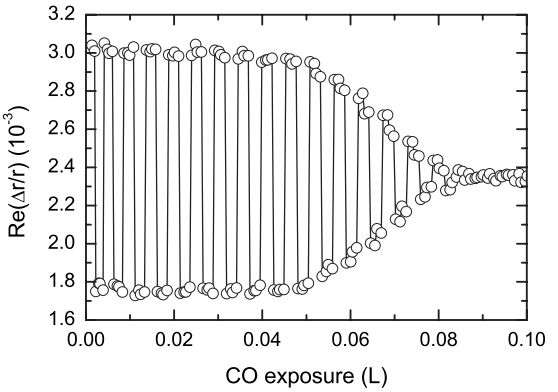

FIG. 3. RD transient recorded at 3.5 eV during the exposure of CO on a 9.1 ML Ni/Cu(110)-(2×1)O film at 150 K. The magnetization direction is periodically switched between pointing into and out of the surface plane by varying the external magnetic field between +1.9 and −1.9 mT.

of the Ni/Cu(110)-(2×1)O system close to the critical coverage can be seen in the influence of CO adsorption on the magnetic properties. First we want to mention that, in contrast to the case of Ni/Cu(001),[1] no SRT back to in-plane magnetization has been found upon decreasing the temperature down to 50 K for a 9.2 ML thick Ni film. On the other hand, CO adsorption at 150 K (i.e., below the CO desorption temperature on oxygen covered Ni) induces a completely reversible SRT for 9–13 ML thick Ni films. As an example, Fig. 3 shows the RD transient recorded during the exposure of a 9.1 ML thick Ni film to a CO partial pressure of 5.5×10^{-10} mbar, while periodically switching the orientation of the applied magnetic field. The amplitude of the magnetic contribution and, hence, the out-of-plane magnetization stays constant up to a critical CO exposure of 0.05 L and is then rapidly quenched to zero within an additional CO exposure of 0.03 L. From this we conclude that, in contrast to the Ni/Cu(001) system, CO adsorption increases the magnetic anisotropy energy[7] of the Ni/Cu(110)-(2×1)O system and is able to switch the magnetization back to in-plane. Additional measurements indicate that the CO adsorption saturates at 2 L. The total CO exposure required for quenching the out-of-plane magnetization linearly increases with the Ni film thickness, whereas the actual reversal of the magnetization direction always occurs within a narrow exposure range of 0.1–0.2 L. For Ni films thicker than 13 ML, CO still has a significant effect on the shape of the hysteresis curve and the coercive field but the Ni film preserves its out-of-plane magnetization. In all cases, the magnetic properties of the clean sample are fully recovered upon thermal desorption of CO.

In summary, we have shown that for Ni films grown at 300 K on the Cu(110)-(2×1)O surface a SRT from in-plane to out-of-plane magnetization occurs at the formation of a contiguous ninth Ni monolayer within a coverage range below 0.1 ML. Within the resolution of our experiments we did not find coexistence of in-plane and out-of-plane magnetism. The previously reported partial out-of-plane magnetization over a coverage range of more than 0.5 ML (Ref. 12) might be due to a different growth morphology or a more

inhomogeneous deposition rate across the sample surface. Finally, we provide evidence for a CO induced, completely reversible SRT back to in-plane magnetization in the Ni/Cu(110)-(2×1)O system.

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[1] M. Farle, W. Platow, A. N. Anisimov, P. Poulopoulos, and K. Baberschke, Phys. Rev. B **56**, 5100 (1997).
[2] P. J. Jensen and K. H. Bennemann, Surf. Sci. Rep. **61**, 129 (2006).
[3] B. Schulz and K. Baberschke, Phys. Rev. B **50**, 13467 (1994).
[4] D. Sander, Rep. Prog. Phys. **62**, 809 (1999).
[5] D. Sander, W. Pan, S. Ouazi, J. Kirschner, W. Meyer, M. Krause, S. Müller, L. Hammer, and K. Heinz, Phys. Rev. Lett. **93**, 247203 (2004).
[6] R. Vollmer, Th. Gutjahr-Löser, J. Kirschner, S. van Dijken, and B. Poelsema, Phys. Rev. B **60**, 6277 (1999).
[7] S. van Dijken, R. Vollmer, B. Poelsema, and J. Kirschner, J. Magn. Magn. Mater. **210**, 316 (2000).
[8] J. Hong, R. Q. Wu, J. Lindner, E. Kosubek, and K. Baberschke, Phys. Rev. Lett. **92**, 147202 (2004).
[9] R. Nünthel, T. Gleitsmann, P. Poulopoulos, A. Scherz, J. Lindner, E. Kosubek, Ch. Litwinski, Z. Li, H. Wende, K. Baberschke, S. Stolbov, and T. S. Rahman, Surf. Sci. **531**, 53 (2003).
[10] S. Z. Wu, G. J. Mankey, and R. F. Willis, J. Vac. Sci. Technol. A **13**, 1497 (1995).
[11] M. Sacchi, A. Mirone, and S. Iacobucci, Surf. Sci. **442**, 349 (1999).
[12] Th. Herrmann, K. Lüdge, W. Richter, K. G. Georgarakis, P. Poulopoulos, R. Nünthel, J. Lindner, M. Wahl, and N. Esser, Phys. Rev. B **73**, 134408 (2006).
[13] R. Nünthel, J. Lindner, P. Poulopoulos, and K. Baberschke, Surf. Sci. **566-568**, 100 (2004).
[14] C. Sorg, N. Ponpandian, A. Scherz, H. Wende, R. Nünthel, T. Gleitsmann, and K. Baberschke, Surf. Sci. **565**, 197 (2004).
[15] M. Wahl, Th. Herrmann, N. Esser, and W. Richter, Phys. Status Solidi C **0**, 3002 (2003).
[16] P. Weightman, D. S. Martin, R. J. Cole, and T. Farrell, Rep. Prog. Phys. **68**, 1251 (2005).
[17] L. D. Sun, M. Hohage, and P. Zeppenfeld, Phys. Rev. B **69**, 045407 (2004).
[18] L. D. Sun, M. Hohage, P. Zeppenfeld, R. E. Balderas-Navarro, and K. Hingerl, Phys. Rev. Lett. **90**, 106104 (2003).
[19] L. D. Sun, M. Hohage, P. Zeppenfeld, R. E. Balderas-Navarro, and K. Hingerl, Phys. Rev. Lett. **96**, 016105 (2006).