In-plane spin reorientation transition in Fe/NiO bilayers on Ag(1,1,10)

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Magnetic properties of Fe/NiO bilayers grown on a (1,1,10) vicinal surface of Ag(001) were studied by magnetooptical Kerr effect (MOKE) and x-ray magnetic linear dichroism (XMLD). The orientation of the antiferromagnetically (AFM) aligned spins of NiO films shows an in-plane spin-reorientation transition (SRT) from parallel to perpendicular to the steps with increasing NiO thickness. Two in-plane SRTs of Fe moments are found in dependence of the NiO thickness. The first SRT of the Fe magnetization from parallel to perpendicular to the steps is observed at NiO thickness where antiferromagnetic order is found, the second at the NiO thickness of the SRT in the NiO film. Also, the in-plane SRT of Fe spins driven by a temperature increase is observed. The perpendicular coupling between the Fe and NiO spins is proven. The induced uniaxial anisotropy energy is estimated to be 0.12 erg/cm².

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I. INTRODUCTION

Magnetic coupling at interfaces allows the tailoring of magnetic properties in magnetic heterostructures, such as ferromagnetic/antiferromagnetic [(FM)/(AFM)] bilayers1,2 and FM/ferroelectric bilayers.3,4 The exchange bias effect in FM/AFM bilayers has attracted much interest for decades, motivated by possible applications in magnetoelectronic devices. The microscopic mechanism responsible for this phenomenon has not yet been fully identified due to difficulties in directly detecting the AFM spin configuration. The existing models explaining the exchange bias effect usually assume an uncompensated AFM surface and a collinear FM/AFM exchange coupling at the interface. However, theoretical calculations for compensated FM/AFM interfaces show that the AFM and FM spins may be coupled orthogonally and that they generate an effective uniaxial anisotropy $K_{xy}$ in the FM layer—just as in our case.5,6

An in-plane SRT driven by perpendicular coupling has been theoretically predicted for temperatures close to the ordering temperature of the AFM layer.7-10 Such perpendicular coupling and in-plane SRT have been observed experimentally in FM/AFM single crystalline systems such as Fe/FeF₂, Fe/stepped-Cr(001), or Fe/MnPd.11-13

In order to understand the exchange coupling in FM/AFM bilayers, it would be very helpful to detect the spin configuration of both FM and AFM films. NiO films are considered an ideal AFM material for studying the magnetic exchange interaction in AFM/FM systems since the antiferromagnetic properties of NiO can be determined by XMLD.14,15

In bulk NiO, the (001) surface is a fully compensated AFM surface; the spins are aligned along the (112) directions with 24 possible orientations. For 0.9 nm thick Fe film grown on a bulk NiO(001) single crystalline surface, the Fe spins were found to be oriented in the sample plane almost perpendicular to the easy axis of bulk NiO.16 For Co films grown on a bulk NiO(001) single crystalline surface, the NiO AFM spins were found to reorient in the sample plane in parallel to the Co spins’ direction.17 However, this interpretation was recently revised by a careful analysis of the NiO XMLD signal showing that Co spins being oriented perpendicular to the NiO spins.18

The AFM spin configuration in NiO films can be very different than in the bulk. The XMLD experiment on NiO film shows that the Ni²⁺ spins lie in the film plane if grown on Ag(001),19 but are normal to the film plane if grown on MgO(001).20 This is due to a different strain effect in NiO films in both cases.19 For a NiO film grown on a bulk Fe(001) surface, the NiO easy magnetization axis was reported to rotate in the sample plane from perpendicular to parallel to the Fe magnetization when increasing the NiO thickness.21

Thus it would be worth examining the coupling between an FM film and the NiO film, which may be different than the coupling for the FM film grown on the bulk NiO substrate.

None of the previous studies on exchange coupling was performed by controlling the AFM spin orientation. We discovered that atomic steps can induce an in-plane anisotropy not only in FM films,22,23 but also in AFM films.24,25 Especially, for NiO films grown at 200°C on the vicinal surface of Ag(001) with steps parallel to [110] the NiO AFM spins exhibit an in-plane SRT from parallel to perpendicular to the steps with increasing NiO thickness.25 This means that the NiO AFM spin orientation can be controlled by the film thickness. So, if growing an FM layer on top of a NiO layer, the exchange coupling can be further studied with the tuned AFM magnetic anisotropy, which may provide a better insight into the magnetism of exchange coupling.

In this contribution we report on the magnetic properties of the Fe/NiO/Ag(1,1,10) system, exploring the exchange coupling between Fe and NiO moments. We first prove that the AFM spins of NiO film grown by pulsed laser deposition (PLD) also show an in-plane SRT from parallel to perpendicular to the steps. In particular, we discuss two SRTs observed as a function of NiO thickness, and the temperature-driven in-plane SRT of Fe which is also observed.
II. EXPERIMENT

The experiments were performed in a multichamber ultra-high vacuum (UHV) system. An Ag(1,1,10) substrate with a vicinal angle of 8° was prepared with cycles of 1 keV Ar ion sputtering and subsequent annealing at 600°C. The sharp double-splitting diffraction spots were formed in low-energy electron diffraction (LEED), as shown in Fig. 1(a). Such splitting of LEED spots has been proven to be proportional to the step density.26 Here, the incident electron beam is oriented along the [001] direction, but 8° off normal to the surface, so in the LEED pattern the light from the filament can be clearly seen as marked by the circle inside. Using scanning tunneling microscopy (STM), we can observe nearly equidistant and regular monatomic steps along the [110] direction from a clean Ag(1,1,10) surface.

The NiO film was prepared by PLD from a NiO target at a growth temperature of 200°C at an O2 pressure of 1 × 10⁻⁶ Torr, similar to the growth conditions of NiO films grown reactively in an O2 atmosphere.27 Then the NiO film was annealed at 300°C for 30 min to improve the surface morphology. LEED indicates an epitaxial growth of the high-quality NiO films. The LEED spots are visible for NiO thicknesses at least up to 10 nm as shown in Fig. 1(b)–1(d). However, the elongated shape of the LEED spots shows that the atomic steps on the NiO surface keep their orientation, but the step density exhibits larger fluctuations than does the step density on the Ag(1,1,10) substrate. The LEED patterns still show clear diffraction spots for thicker films up to 10 nm. The NiO film was grown as a wedge shape with a slope of ~1 nm/mm. This allowed us to systematically study the thickness dependence of the magnetic properties of a NiO film for one sample, i.e., with the same growth conditions for all the investigated thicknesses. In order to study the exchange coupling between NiO and Fe, a 2 nm Fe film was grown on top of NiO by molecular beam epitaxy (MBE) at room temperature (RT). The Fe growth rate was calibrated using reflection high energy electron diffraction (RHEED) intensity oscillations prior to the film deposition. The magnetic properties were studied in situ by MOKE without any field cooling treatment of the samples. The incident laser beam (wavelength 670 nm, diameter 0.2 mm) is s-polarized with a 30° incident angle. Only Kerr ellipticity measurements will be presented and discussed in this paper.

III. RESULTS AND DISCUSSION

Previously, for NiO films reactively grown in an O2 atmosphere on Ag(1,1,8),25 the NiO AFM spins were found to be oriented parallel to the steps up to a thickness of 3.5 nm, and perpendicular to the steps for thicker NiO films. In order to verify the orientation of the AFM spins in the NiO film grown by PLD, we first carried out XMLD measurements of the Au/NiO/Ag(1,1,10) system. The experiments were performed in the UES6/2-PM2 beam line at BESSY in Berlin. A 10 monolayers (ML) Au capping layer was grown to protect the NiO film from contamination. The sample was kept in a high vacuum while being transferred from the growth chamber in Halle to the XMLD chamber at BESSY. Figure 2 shows the x-ray adsorption spectra detected at RT with the incident x-ray normal to the NiO films of different thicknesses, for the polarization direction both parallel and perpendicular to the steps. Here the XAS spectra are normalized to the lower-energy peak. A clear XMLD signal demonstrates an...
AFM order in the NiO film. The Ni\textsuperscript{2+} L\textsubscript{2} spectrum of the NiO film consists of two absorption peaks. The relative intensities of these two peaks can be used to determine the spin orientation in the NiO film.\textsuperscript{14,15,18} For the 1.7 nm thick NiO film, the peak of relative intensity larger for E\textparallel steps exhibits smaller intensity for E\perpendicular steps. The relative peak intensity of the Ni L\textsubscript{2} doublet are clearly reversed for the 3.3 nm thick NiO film. A recent study by Arenholz et al.\textsuperscript{18} states that if Ni\textsuperscript{2+} spins are parallel to the [110] axis, then the higher-energy peak of the L\textsubscript{2} doublet should reach its minimum value when the x-ray polarization vector is parallel to the Ni spins direction. Therefore, we can conclude that the average Ni\textsuperscript{2+} spins in our NiO/Ag(1,1,10) system are parallel to the steps for 1.7 nm of NiO and perpendicular to the steps for 3.3 nm of NiO. This result is consistent with our previous report on NiO films grown by evaporating Ni onto Ag(1,1,8) in an O\textsubscript{2} atmosphere.\textsuperscript{25} The reorientation of the AFM spins of the NiO film can be attributed to structural relaxation of the NiO films at a thickness of d\textsubscript{NiO} ∼ 2 nm.\textsuperscript{28} Such a relaxation thickness may vary with different growth methods.

The magnetic properties of the Fe/NiO/Ag(1,1,10) system were studied by longitudinal MOKE. Figure 3 shows magnetic hysteresis loops measured at RT for 2 nm of Fe grown on top of NiO films of different thicknesses. According to the results of Ref. 32, an exchange bias effect is not expected at RT for NiO film thinner than 25 ML. The external magnetic field was applied either parallel or perpendicular to the steps. Double split loops were measured for the magnetic field perpendicular to the steps [as shown in Fig. 3(a)] and rectangular easy-axis loops were measured for the external field parallel to the steps [as shown in Fig. 3(b)]. Such double-split loops are not surprising since it is known that the atomic steps can induce an in-plane uniaxial anisotropy (in addition to a fourfold anisotropy) with the easy axis parallel to the step direction, observed previously, e.g., for uncovered Fe films grown on an Ag(1,1,10) clean surface.\textsuperscript{22} The double split loops can be characterized by the shift field H\textsubscript{s}, which is related to the anisotropy modification due to the steps.\textsuperscript{29,30} For the Fe(2 nm)/NiO/Ag(1,1,10) system, it can be seen from Fig. 3 that H\textsubscript{s} increases from 30 Oe to 280 Oe when the thickness of NiO increases from 0 to 0.4 nm, i.e., to a thickness at which the Fe/NiO interface is fully developed. H\textsubscript{s} remains almost unchanged (with a value of ∼280 Oe) for 0.4 nm < d\textsubscript{NiO} < 0.8 nm. In NiO films thicker than 1 nm, the easy magnetization axis of Fe switches by 90°, i.e., to a direction perpendicular to the steps. Consequently, the hysteresis loops for H\perpendicular steps show a rectangular shape [Fig. 3(e)], whereas double-split hard-axis loops appear for H\parallel steps [Fig. 3(f)]. With a further increase of the NiO thickness for d\textsubscript{NiO} > 2.8 nm, the alignment of the Fe easy magnetization axis switches again from perpendicular to parallel to the steps. The data show that for Fe/NiO/Ag(1,1,10) an in-plane SRT occurs twice at two different NiO thicknesses.

Usually, the anisotropy modification due to steps is discussed as the involvement of interfacial and volume contributions.\textsuperscript{22} A strong dependence of H\textsubscript{s} on d\textsubscript{NiO} for thin NiO films in comparison to what has been observed for the Fe/Ag(1,1,10) system can be attributed to the interfacial contribution to the anisotropy. Obviously, the interfacial anisotropy changes by inserting the NiO layer in between the Ag(1,1,10) substrate and the Fe film. The interfacial contribution is very sensitive to the atomic configuration and can be strongly influenced by any surface/interface modification. For example, after covering the Fe/Ag(1,1,10) system with Au,\textsuperscript{22} the surface contribution to the step-induced anisotropy modification can increase by a factor of 2. Therefore, it is very likely that the step-induced anisotropy may change by replacing the Fe/Ag interface with an Fe/NiO interface.

After having clarified those points, we discuss the origin of two in-plane SRTs observed at two different NiO thicknesses. When the NiO film is very thin, it is in a paramagnetic state and the step-induced anisotropy modification of the Fe film is dominant in the total anisotropy. Thus the Fe easy axis is oriented along the steps. It is well known that the Néel temperature of an AFM layer increases with film thickness. Therefore, the AFM state of NiO can be established at RT at its certain thickness. The reported critical thickness of NiO at RT is ∼1.2 nm,\textsuperscript{25,31} which is close to the NiO thickness at which the first SRT in the Fe film is observed. Since the step-induced anisotropy should align the easy magnetization axis of Fe along the steps, its switching into a direction perpendicular to the steps at d\textsubscript{NiO} ∼ 1 nm can only be explained by the appearance of an AFM order at RT in the NiO film of this thickness. Since the XMLD experiment already showed that NiO spins above a thickness of 1 nm are parallel to the steps,\textsuperscript{25} one can
conclude that the Fe and NiO moments are coupled together perpendicularly. The XMLD measurements have shown that the NiO spins switch from parallel to perpendicular to the steps at a thickness of \( d_{\text{NiO}} \sim 2.8 \text{ nm} \) due to lattice relaxation. Thus, due to perpendicular coupling between the spins of Fe and NiO, the Fe easy magnetization axis switches again to an alignment along the steps.

Figure 4 shows the phase diagram for the easy magnetization axis of the Fe film with respect to the steps as a function of temperature and NiO thickness. The NiO thicknesses of the in-plane SRTs in the Fe film were determined at three different temperatures: 5 K, 300 K, and 380 K from the hysteresis loops measured for varying thicknesses of NiO. The NiO thickness of the first SRT in Fe corresponds to the thickness at which the AFM order is established, thus it is expected to depend on temperature. Consequently, the thickness of the first SRT in the Fe film is strongly temperature dependent. The second SRT in the Fe film occurs due to the SRT in the AFM NiO film, which is attributed to the structural relaxation of the NiO films. Because of that the second SRT transition thickness has very weak temperature dependence.

To further prove whether the first SRT in the Fe film originates from the AFM order in the NiO film, temperature-dependent MOKE measurements were performed on Fe/NiO/Ag(1,1,10) with an NiO film of \( d_{\text{NiO}} = 1.8 \text{ nm} \) thickness. At RT, the 1.8 nm thick NiO film is AFM with NiO moments oriented along the steps, so the perpendicular coupling aligns the Fe moment perpendicularly to the steps. With increasing temperature above the NiO film's Neel temperature, the step-induced anisotropy aligns the magnetization of the Fe film parallel to the steps. Therefore, the magnetic hysteresis loop is expected to change from a rectangular to a double split hard-axis loop if the field is applied perpendicular to the steps. This indeed happens as shown in Fig. 5. The Fe/NiO interface was proven to be chemically stable up to 540 K. Consequently, after applying no more than 430 K (Fig. 5) the hysteresis loop can change back to a rectangular loop after cooling down to RT, i.e., the SRT is reversible.

The exchange coupling energy between the Fe layer and the NiO layer can be calculated quantitatively from the experimental results if assuming that the uniaxial anisotropy \( K_\alpha \) and the shift field \( H_s \) are proportional, i.e., \( K_\alpha \sim M_{Fe}d_{Fe}H_s \). For the Fe(2 nm)/NiO(1.8 nm)/Ag(1,1,10) system, the shift field \( H_s \) is \( \sim 130 \text{ Oe} \) at RT, but increases to 230 Oe when the easy axis switches to being parallel to the steps at 430 K. So by ignoring a possible small variation of the step-induced uniaxial anisotropy due to the temperature increase from RT to 430 K, the anisotropy energy \( K_{sf} \) induced by the exchange coupling between the Fe film and the AFM NiO film can be estimated to be \( \delta K_e = K_{sf} \sim 0.12 \text{ erg/cm}^2 \). This value is higher than the unidirectional anisotropy reported for the single-crystalline Fe/NiO(001) system. In those studies, the maximum exchange bias field of 220 Oe was obtained for a 10 ML thick Fe film indicating the unidirectional anisotropy of \( \sim 0.05 \text{ erg/cm}^2 \). The interlayer exchange coupling between neighboring FM and AFM spins across the interface can be estimated as \( J^2_{int} = K_{sf}J_{AFM} \), where \( J_{AFM} \) is the exchange coupling in the AFM system. In theoretical models, such interlayer exchange coupling is responsible for the exchange bias effect on an uncompensated AFM surface. We therefore believe that our result can be useful for further studies regarding the strength of \( J_{int} \) at Fe/NiO interfaces, which is usually difficult to determine experimentally.

In summary, we studied the magnetic properties of Fe/NiO bilayers grown on Ag(1,1,10). Using the XMLD technique we discovered that the easy axis of the NiO AFM spins for thin NiO films is aligned along the steps, and for thicker films perpendicular to the steps. The easy magnetization axis of the Fe film in the Fe/NiO/Ag(1,1,10) system switches from parallel to perpendicular to the steps for \( d_{\text{NiO}} \sim 1.0 \text{ nm} \), and back to the step direction for \( d_{\text{NiO}} > 2.8 \text{ nm} \). The two in-plane SRTs are driven by perpendicular coupling between the Fe and NiO spins at the Fe/NiO interface. The anisotropy energy induced by the exchange coupling is estimated as \( \sim 0.12 \text{ erg/cm}^2 \).
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