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Surface antiferromagnetism of NiO studied by photoemission microscopy

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Abstract

Antiferromagnetic domains at surfaces of NiO single crystals were studied by polarization dependent soft X-ray absorption at the Ni2p absorption edges. Spatially resolved information is obtained by monitoring the absorption via the total yield of electrons in a photoemission electron microscope. On the (100) surface, stripe shaped domains are found with widths of the order of a few 10 μm , extending over 100s of μm . The polarization dependence shows a sign change of the contrast for every 90° of rotation, as expected for the antiferromagnetic twin domains with the major portion of the antiferromagnetic axis within the plane of the surface. The domains are not affected by surface topography. This suggests that the formation of domains is governed by defects in the bulk, which lead to nucleation of domains at specific sites. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Antiferromagnetic order occurs for a large number of transition metal and rare earth materials, both metallic and non-metallic. As for other collective phenomena, one may expect that the presence of a surface severely modifies the antiferromagnetic order. Such modifications may manifest themselves in the transition temperature, the magnetic anisotropy, the type of antiferromagnetic order, or the magnitude of the magnetic moments. For ferromagnetic surfaces and ultrathin films these phenomena

have been at the focus of attention for a long time, and by now a sizeable body of information has been established concerning modification of ferromagnetic properties at or near to surfaces and interfaces. For antiferromagnetic (abbreviated AF) surfaces and films, our knowledge is much more restricted, partially because the experimental methods for the study of AF surfaces are not as highly developed. The other aspect which probably made antiferromagnetic materials less attractive for experimental investigations is the lack of technical applications. This aspect has changed dramatically in recent years with the development of novel magnetic devices, such as read heads for data storage systems or other magnetic sensors based on magnetoresistance phenomena. Even though the key components in these systems

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are still ferromagnetic films, antiferromagnetic layers are nearly always incorporated with the purpose of shifting the hysteresis loop of one of the the ferromagnetic layers away from being centered symmetrically around zero magnetic field. This phenomenon is known as exchange bias, and is caused by the exchange coupling of the magnetic moments of the ferromagnet and the antiferromagnet at their interface. Clearly, a detailed knowledge of the spin and domain structure at the interface is needed to understand and optimize exchange bias, both for the ferro- and the antiferromagnet.

The AF structure of NiO consists of ferromagnetic (111) sheets, stacked in an antiferromagnetic sequence. This leads to four different domains associated with the four (111) planes, each of which can split up further into domains depending on how the spins are oriented within the (111) planes. Because of the threefold symmetry of the (111) planes, three such domains are possible, leading to twelve domains overall (omitting spin reversal). Within a single domain, the spins are collinear. Domains can be characterized by a so-called antiferromagnetic axis, which is given by a unit vector parallel to the *difference* the two sublattice magnetizations. For a collinear antiferromagnet, such as NiO, the AF axis is parallel to one of the sublattice magnetizations.

The prime method for investigating antiferromagnetism is neutron scattering, where the increased magnetic unit cell leads to additional reflections [1–3]. This technique provides information on the symmetry of the magnetic order. Also optical techniques have been used [4,5]. The detailed spin orientation is more difficult to extract. Neutron topography on crystals subjected to a treatment to increase the domain size showed that the spins are aligned along [112] directions lying within the (111) planes [3]. However, in a recent study of first order magnetic Raman scattering on NiO it was found that the polarization dependence cannot be described by the generally accepted antisymmetric scattering tensor [6]. Inclusion of higher order terms in the magneto-optic coupling can explain the observed polarization dependence, but is not consistent with the [112] spin alignment. Therefore, an independent and if possible more direct probe of the spin orientation of NiO is desirable.

It is well known that soft X-ray absorption spectra

of transition metal compounds show rich structure arising from Coulomb and exchange interaction between the localized core hole and the valence electrons, which in such compounds are also highly localized. From a detailed analysis of the NiO spectrum it is expected that the line shape depends on the relative orientation of (linear) light polarization and the magnetic moments, which for a collinear antiferromagnet coincide with the AF axis. The polarization dependence was confirmed experimentally on thin NiO films which by their thin film nature had a preferred orientation of the magnetic moments perpendicular to the film plane. Photoemission microscopy experiments using this effect on ultrathin NiO films grown on Ag(100) showed lateral changes of the relative abundance of different domains, while the domains themselves were below the spatial resolution limit [8]. For epitaxial films on MgO, Stöhr et al. [9] observed stripe-shaped domains related to defect structures. Here we report on the observation of intrinsic antiferromagnetic domains at NiO surfaces by photoemission microscopy, using soft X-ray linear magnetic dichroism as a contrast mechanism.

2. Experiment

The experiments were performed using synchrotron radiation from the elliptical undulator UE 56/1 at the new BESSY II storage ring in Berlin [10], as well as undulator radiation from the BW 3 beamline at Hasylab in Hamburg [11–14]. In the soft X-ray regime, absorption is conveniently measured by monitoring the total yield of electrons. To obtain spatial resolution, we measure the yield by an electrostatic photoemission electron microscope (PEEM 350 by Staib Instruments [15,16]). The microscope is mounted in a UHV chamber with its electron optical axis at 75° to the synchrotron beam, such that the light incidence onto the sample is under 15° measured to the surface. At the BESSY beamline, the light polarization can be changed between s-polarized (electric field lying in the sample surface) and p-polarized (electric field at 75° to the surface), while only s-polarization is available at BW 3.

Sample preparation is carried out in a separate UHV chamber, equipped with LEED, sputter gun and

deposition sources. The sample load lock system comprises a cleaving stage, and allows also for UHV transfer between the two chambers [17]. The samples were single crystals cleaved in air or in UHV. Surfaces cleaved in UHV often exhibited sharp features which lead to substantial field emission when exposed to the electric field of $30\text{--}40\text{ kV cm}^{-1}$ under the photoemission microscope. This problem is less severe or absent for surfaces cleaved in air, and — depending on the duration of exposure to atmosphere — short sputtering after insertion into vacuum. The overall domain patterns were similar for in situ and ex situ cleaved surfaces.

3. Results

To image antiferromagnetic domains, we make use of linear dichroism in soft X-ray absorption at the $\text{Ni}2p_{1/2}$ edge. This phenomenon leads to a change of the absorption spectrum when the light polarization is changed from being parallel to perpendicular to the AF axis. It arises from the selection rule for the magnetic quantum number $\Delta m = 0$ or ± 1 , relevant for these two situations [7]. In principle, also the $2p_{3/2}$ edge is expected to show dichroism. However, the mean absorption cross section varies strongly, such that at this edge the contrast occurs between features in the absorption spectrum which differ strongly in intensity. At the $j=1/2$ threshold, the features giving the contrast have similar intensity, which makes this part of the absorption spectrum more suitable for domain imaging. The $\text{Ni}2p_{1/2}$ absorption spectrum shows two peaks at 871.2 and 872.2 eV, whose relative intensity is different for light polarization parallel or perpendicular to the magnetic moment. For domain imaging, the intensity ratio is taken pixel by pixel of two images taken with the photon energy tuned to the $2p_{1/2}$ absorption features [8,9,18].

Fig. 1 shows two domain images taken from the same area on a $\text{NiO}(100)$ surface. The images show a grey scale coding according to $256 * [(I(x,y) @ 871.2\text{ eV}) / (I(x,y) @ 872.2\text{ eV}) - 1]$, where $I(x,y)$ are the intensities as a function of position at the photon energies given. The light was s-polarized, incident at 15° to the surface. The contrast shown Fig. 1 disappears reversibly on heating the sample above

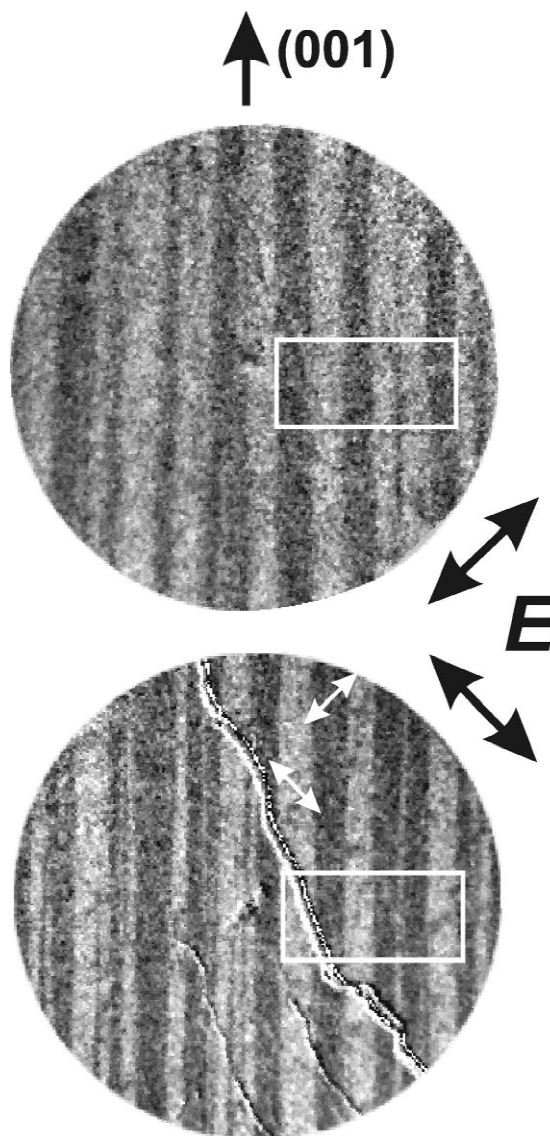


Fig. 1. Photoemission micrographs showing antiferromagnetic stripe domains parallel to the [001] direction in the (100) NiO surface; the field of view is $250\ \mu\text{m}$ in diameter. The light is s-polarized, with the direction of the electric field indicated by the double headed arrows. Rotation of the light polarization (by rotating the sample) by 90° between upper and lower panel leads to contrast reversal. This can be seen well in the areas delineated by white rectangles. The magnetic moment directions are schematically indicated by the double headed white arrows in the lower panel.

525 K, which is the Néel temperature of NiO. Therefore, the observed contrast can unambiguously be attributed to the antiferromagnetic ground state of NiO. Domains with a large or small projection of the AF axis on the electric field show up as bright or dark stripes in the image. By rotating the sample about its surface normal, the orientation of the electric field is changed relative to the crystal axes, and therefore also with respect to the orientation of magnetic moments (AF axis) in the crystal. As shown in Fig. 1, we observe a reversal of contrast between AF domains for rotation of the light polarization relative to the crystal by 90° . On rotating the sample, some refocussing is required, which leads to a slightly different size of the field of view in the two images. In addition to the stripe domains, the lower image shows some irregular lines. These are caused by topographical features on the surface, which are not completely removed on subtraction or division of the two images [19].

The stripe domains observed here have widths of the order of a few $10\ \mu\text{m}$, and extend over 100s of μm . This is much larger than the intrinsic domain sizes inferred indirectly for epitaxial films [8,9]. The main difference to those studies is that here a massive single crystal was used. As no special treatment had been applied to increase the domain size, the observed domain size can be regarded as a typical one for these crystals. Since there is no stray field contribution to the free energy, the domain topography must be determined by other properties. The most likely one appears to be magnetostriction, or, as it is often termed for antiferromagnets, exchange striction. The antiferromagnetic structure of NiO consists of ferromagnetic (111) sheets, stacked in antiferromagnetic sequence. In the antiferromagnetic state, the spacing between these (111) planes is slightly reduced from the spacing of the undistorted cubic NaCl structure by magnetostriction. Conversely, mechanical stresses will influence the antiferromagnetic structure. We propose that defects incorporated into the crystal during the growth process lead to stresses, which in turn govern the domain structure of a massive antiferromagnetic crystal. As can be seen in Fig. 1, large defects *at the surface*, like ridges, etc., do not influence the domain structure for the example shown. This indicates that the domain pattern observed in our experiment represents the surface termination of the bulk domain structure.

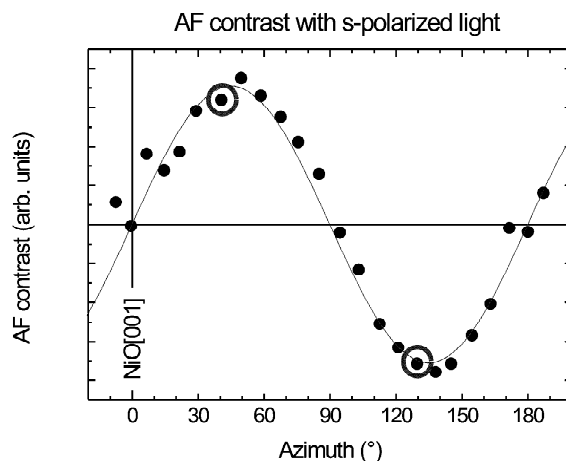


Fig. 2. Antiferromagnetic contrast (for definition see text) on NiO (100) excited at the $\text{Ni}2p_{1/2}$ threshold as a function of azimuthal sample orientation. The orientations at which the images in Fig. 1 were taken are marked by large circles. The contrast vanishes and changes sign every 90° rotation.

Fig. 2 shows the change of contrast between antiferromagnetic domains for a sample rotation by 180° . The two sample orientations which are represented in Fig. 1 are marked by circles. As these positions are not 90° apart, the contrast is slightly different in the two images of Fig. 1. Nevertheless, it can be seen that the AF contrast is close to maximum for the sample orientations shown in Fig. 1. The maxima in the contrast occur for angles of 45° between electric field of the light (light polarization) and the [001] or [010] directions in the crystal surface. This means that the directions of magnetic moments AF axis are roughly at 45° to the stripes, as indicated by the arrows labelled ' μ ' in Fig. 1.

For applications, e.g. in novel magnetic devices based on spin dependent transport, ultrathin antiferromagnetic films are used to tailor the hysteresis loop of a ferromagnetic film to specific requirements. Specifically, the exchange coupling at the interface between the ferro- and antiferromagnetic films induces a shift of the ferromagnetic hysteresis loop away from being centered symmetrically around zero field. This so-called exchange bias leads to different coercive fields for the two magnetization directions of the film in direct contact with the antiferromagnet. For a layered antiferromagnet which can be thought of an antiferromagnetic stacking of ferromagnetic planes, one may expect that a surface which co-

incides with these planes should behave like a ferromagnetic surface. Nevertheless, these ferromagnetic moments will not be affected by external fields because of the large antiferromagnetic exchange interaction with the deeper layers of the antiferromagnet. This suggests that AF surfaces which on the basis of the bulk AF structure should exhibit a net magnetization should be particularly effective for exchange bias. To address this issue, we investigated ultrathin films of NiO grown on Au(111). The (111) surface of the NaCl structure is polar, and therefore does not occur in unreconstructed form. We grew NiO films epitaxially on the Au(111) surface, using the procedures established by Neddermeyer and co-workers [20], with a film thickness of 30 monolayers. Deposition of Ni in an atmosphere of about 10^{-6} mbar O_2 leads to the formation of NiO. X-ray absorption spectra of such films showed all the features characteristic for NiO. Due to the surface orientation, different domain patterns may be expected from those observed on the (100) surface. However, microscopic images taken with the photon energy tuned to the $Ni2p_{1/2}$ excitation threshold did not show any domains. Absorption data taken with different azimuthal sample orientations did, however, show changes of the absorption spectra. Fig. 3 shows such a series of $Ni2p_{1/2}$ absorption spectra taken by total yield, i.e. without spatial resolution, from the NiO/Au(111) sample. The change of the absorption spectrum suggests that there is a preferred orientation of AF axis, or even a single or very large domain, of a size comparable to or larger than the size of the

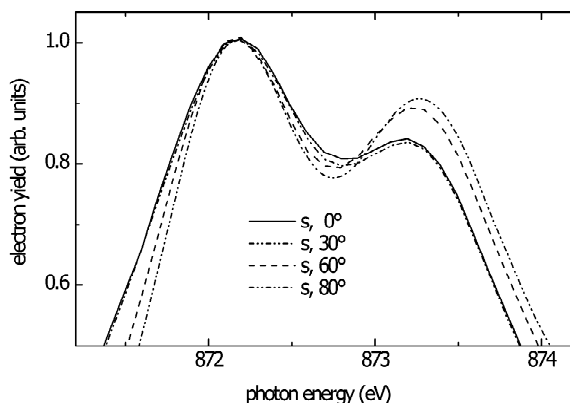


Fig. 3. Series of $Ni2p_{1/2}$ absorption spectra for a NiO film grown on Au(111) for different azimuthal orientations.

soft X-ray beam. Further studies of epitaxial films on Au(111) are being carried out.

4. Summary and outlook

The new approach for imaging antiferromagnetic domains opens the way to a wide range of issues in connection with antiferromagnetic surfaces. An important feature which makes this technique unique among the methods for antiferromagnetic materials is the high surface sensitivity. The combination with chemical selectivity resulting from core level excitation makes this technique capable of detecting ultrathin films in the monolayer range. The other remarkable feature is the speed: images as those shown here require acquisition times on the order of minutes, compared to typical acquisition times of 20 h for neutron topography. By utilizing the polarization dependence, spin structures at surfaces can be determined. Also the investigation of the critical behaviour on approaching the Néel temperature is easily possible.

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