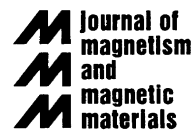




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Imaging of antiferromagnetic domains at the NiO(1 0 0) surface by linear dichroism in near UV photoemission microscopy

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Abstract

Antiferromagnetic domains at the NiO(100) single-crystal surface are imaged by photoemission microscopy excited by linearly polarized near UV radiation from a mercury vapour light source. The contrast is observed by subtracting images excited by s- and p-polarized light. This contrast is compared with the contrast from X-ray magnetic linear dichroism (XMLD). Here the XMLD contrast is gained by subtracting two images excited by the same polarization but different photon energies at the Ni2p core excitation. © 2001 Elsevier Science B.V. All rights reserved.

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The concept of magnetic domains was proposed by Weiss to explain the empirical observation that a piece of iron can appear non-magnetic at room temperature, well below the Curie temperature [1]. In the demagnetized state, parts of the sample are magnetized in different directions, such that the macroscopic magnetization vanishes. The driving force for the formation of domains is the minimization of the magnetic stray field energy, which is balanced by the energy cost of forming domain walls. Apart from ferromagnetism, long-range magnetic order may also – and probably even more abundantly – occur as antiferromagnetism, and this state is also connected with the occurrence of magnetic domains [2]. In the simplest case, an antiferromagnetic crystal can be described as consisting of two ferromagnetic sublattices with mutually opposite magnetizations. The magnetic order is conveniently described by the difference of the sublattice magnetization vectors, termed antiferromagnetic (AF) vector in the following. Domains in antiferromagnetic crystals may then be characterized by AF vectors pointing in different directions.

Because of the vanishing macroscopic magnetization, there is no stray field associated with a finite antiferromagnetic solid. Therefore, one might expect very large domains in AF materials. On the other hand, since one of the dominating contributions to the energy balance is quenched, the domain pattern will be determined by the delicate balance of the smaller contributions to the total energy, and this may lead to specific domain patterns in antiferromagnetic materials. To identify the important contributions and their connection to the occurrence of various types of domains and domain walls in antiferromagnets, experimental data on AF domain topography for a wide range of AF materials and different crystallographic surfaces are highly desirable. However, because of the vanishing magnetization, magneto-optical methods from which the vast majority of data on ferromagnetic domains is derived, are rendered ineffective since most of the magneto-optical effects are odd functions of the magnetization. But there is one magneto-optical effect, which is an even function of the magnetization, and that is the Voigt effect: it occurs as a change of the optical properties for light polarization either parallel or perpendicular to the magnetization. Phenomenologically, for a cubic system with a magnetic order along the z-axis, it is described by the xx and yy components of the dielectric tensor being different from the zz component. This may be described

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as linear birefringence, or in an absorptive medium linear dichroism.

As for other magneto-optic effects, also linear magnetic dichroism may be much larger in the soft X-ray regime at core level excitation thresholds than for wavelengths in the visible. This is connected to the large spin-orbit interaction of the core levels. In particular, for highly localized systems like oxides, large effects have been predicted [3] and observed [4–6]. The observations are well reproduced by calculations of the polarization dependence of the soft X-ray absorption spectra. The easiest way to measure absorption spectra in the soft X-ray regime is by monitoring the total yield of secondary electrons. Spatially resolved X-ray absorption spectra can be obtained by monitoring the electron yield in a photoemission microscope. Therefore, the combination of linear magnetic dichroism with photoemission microscopy offers an attractive route for imaging AF domains. The power of this technique has been demonstrated recently by a number of reports [7–9].

Circular and linear dichroism in the soft X-ray region are only appreciable for photon energies close to the core level excitation thresholds [10], e.g., the 2p and 3p thresholds. This shows that the off-diagonal elements viz. the difference between the diagonal elements of the dielectric tensor vanishes at energies far from the core level thresholds. From this it might be expected, that there is no dichroism in the threshold photoemission, where the photon energies just exceeds the work function, since for such energies no core levels are involved. However, the occurrence of magneto-optic effects in the visible regime suggests that also for interband transitions spin-orbit effects are present, giving rise to small dichroic effects. Therefore, we searched for dichroic effects which can be used for domain imaging in threshold photoemission of NiO. An analogue effect on ferromagnetic surfaces was reported recently by Marx et al. [11] for threshold photoemission microscopy of Fe. In this report we demonstrate that also in threshold photoemission from antiferromagnetic NiO there is a linear dichroism which can be used for imaging of antiferromagnetic domains. As the spin-orbit coupling is smaller in the valence states than for core states, the effects are expected to be smaller. On the other hand, the narrow energy spread of photoelectrons excited by near UV light is of advantage for achieving higher spatial resolution, and also the possibility to image AF domains independent from synchrotron radiation is attractive.

The apparatus used in this study consists of two UHV chambers, one for the microscope and one for sample preparation. Sample can be introduced via a fast entry load lock. The transfer between load lock and the two chambers is accomplished by a magnetically coupled transfer rod. The photoemission microscope (PEEM 350,

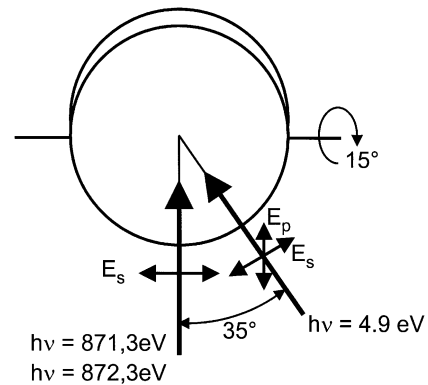


Fig. 1. Experimental set up: Contrast can be achieved by switching between s- and p-polarized light from the Hg arch lamp or by switching between two energies of the synchrotron light. The sample is tilted 15° to the plane defined by the two light beams. The rotation axis is perpendicular to the synchrotron light beam.

manufactured by Staib Instruments [12]) transfers the electrons released from the sample by an electrostatic lens column to a channel plate/screen assembly, from which an enlarged map of the emission probability is recorded in a CCD camera. For excitation of the photoelectrons, we used monochromatic synchrotron radiation in the soft X-ray regime or light of an Hg arc lamp. The experimental set-up allows both photon sources to be used without a change of the geometry between sample and PEEM. The grazing angle was 15° for the synchrotron light and $\sim 12^\circ$ for the Hg arc lamp. The angle between synchrotron light and Hg arch lamp was 35° (Fig. 1). The synchrotron light was provided by the BW3 undulator beamline at HASYLAB, Hamburg. It is linearly polarized, with the electric field lying in the sample plane (s-polarized). The light of the Hg arc lamp has a prominent line at 254 nm wavelength (4.88 eV). Light in the visible range and lower photon energy is of no importance since these photons cannot transfer sufficient energy to the electrons to overcome the work function of 4.3 eV [13]. Between the Hg lamp and the UV window of the chamber, a LiF polarizer was installed to allow for quick changeover between s- and p-polarization.

The images shown in this paper are taken from a NiO(100) single crystal. We used two different methods for the surface preparation. The images in Figs. 2 and 3 were obtained from a NiO-crystal in situ cleaved to expose an uncontaminated (100) surface. For further investigations that are of no concern in the context of this report the surface was decorated with three monolayers CoO. The upper images show the surface topography of the sample. The edges visible in the upper images of Figs. 2 and 3 demonstrate that the images show the same

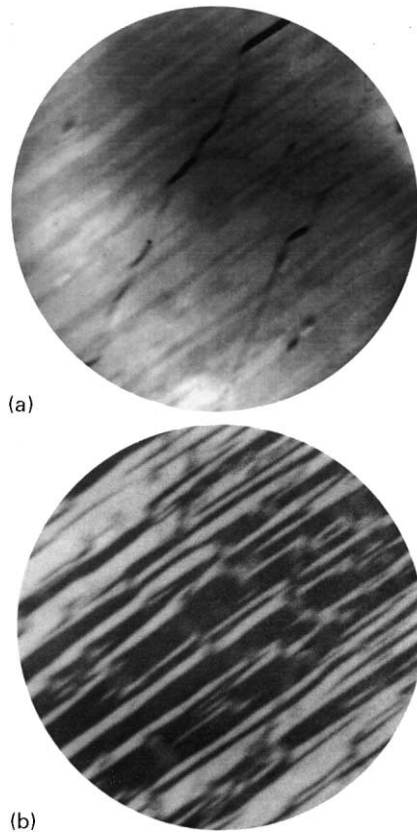


Fig. 2. Photoemission microscopy of a cleaved NiO(100) crystal surface (decorated with CoO). Linearly s-polarized, tuned to the Ni $2p_{1/2}$ threshold, was used for excitation. (a) Topography taken at 871.3 eV. The antiferromagnetic domains are visible in single images excited at the Ni $2p_{1/2}$ energy. (b) Image of the ratio $I(872.3 \text{ eV})/I(871.3 \text{ eV})$, showing antiferromagnetic domains with enhanced contrast. The field of view of all images is $300 \mu\text{m}$.

region. Depending on light incidence, edges on the sample appear darker or brighter than the surrounding. With synchrotron excitation (Fig. 2), the AF contrast on the cleaved surface is large enough that a single image excited at the Ni $2p_{1/2}$ energy shows the AF contrast by itself, as revealed by the stripes running from lower left to upper right, without the subtraction of a reference image or any other processing. The bright and dark stripes visible in the lower image, which shows the ratio of intensities with 871 and 872 eV excitation, show the antiferromagnetic domains with higher contrast. The AF contrast disappears on averaging over the two images. The spins are either (near) parallel or (near) perpendicular to the light polarization. The connection to antiferromagnetic domains was established by the reversible disappearance of this contrast at the Néel temperature [9]. The UV images were obtained using both linear s-

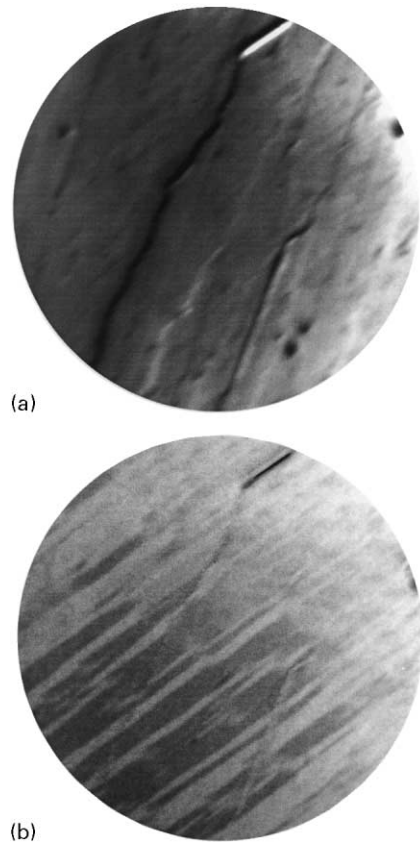


Fig. 3. Photoemission image of the cleaved NiO(100) crystal surface, decorated with CoO. Excitation by Hg arc lamp with prominent wavelength 254 nm. (a) Topography taken with p-polarized light. (b) Contrast $I(E_p)/I(E_s)$ showing the same domain pattern as observed with excitation at the Ni $2p_{1/2}$ threshold, see Fig. 2.

and p-polarizations. The image of the ratio shows the same domain pattern as the image excited at the $2p_{1/2}$ energy. This shows conclusively that the origin of the contrast observed with UV excitation is also the antiferromagnetic domain structure. The contrast observed in these images can be up to 9%, which is not much smaller than the 16% found with the X-ray excitation. On the other hand, for the experimental conditions used in this study the resolution in the UV excited image is better. From the pronounced dependence of the overall electron yield on sample alignment it appears that the probing depth is much smaller for UV excitation than for soft X-ray excitation. This is plausible since the UV photons have a shorter absorption length than the X-rays. The energy dependence of the probing depth and the AF contrast can be used to differentiate between properties

characteristic of the region in immediate vicinity of the surface and the region up to 5 nm below the surface as probed by total yield with X-ray excitation. For this purpose, systematic studies of the probing depths with different excitation sources will be performed.

Analogous results were also obtained for crystals cleaved ex situ, which had been stored in air for long periods. In these cases, the AF contrast was observed after mild sputtering of the surface. Typically, we exposed the sample to $20 \mu\text{A} \cdot 0.5 \text{h}/\text{cm}^2$ Ar ions with 500 eV kinetic energy to remove contamination. Under these conditions, one may expect the surface layer to have an oxygen deficit of about 15% [14]. The observation of AF domains under highly surface sensitive UV excitation shows that this does not strongly modify the AF order in the surface layers.

Optical birefringence has been used in a number of studies [15,16] for the investigation of AF domains in NiO. This shows that also in the visible region of the spectrum spin orbit coupling is sufficient to yield measurable effects, which allow distinction between differently oriented domains. Here we demonstrate that also in the near UV the effects are large enough to allow for imaging of AF domains. The microscopic origin of the dichroism still has to be clarified. By combining threshold excitation with core level excitation, one can make use of different surface sensitivities to further clarify near surface structure of antiferromagnetic transition metal oxides.

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