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Exploring the microscopic origin of exchange bias with photo-electron emission microscopy (PEEM)

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Abstract

It is well known that magnetic exchange coupling across the ferromagnet – antiferromagnet interface results in a unidirectional magnetic anisotropy of the ferromagnetic layer, called exchange bias. Despite large experimental and theoretical efforts, the origin of exchange bias is still controversial, mainly because detection of the interfacial magnetic structure is difficult. We have applied photoelectron emission microscopy (PEEM) on several ferromagnet - antiferromagnet thin-film structures and microscopically imaged the ferromagnetic and the antiferromagnetic structure with high spatial resolution. Taking advantage of the surface sensitivity and elemental specificity of PEEM, the magnetic configuration and critical properties such as the Néel temperature were determined on $LaFeO_3$ and NiO thin films and single crystals. On samples coated with a ferromagnetic layer, we microscopically observe exchange coupling across the interface, causing a clear correspondence of the domain structures in the adjacent ferromagnet and antiferromagnet. Field dependent measurements reveal a strong uniaxial anisotropy in individual ferromagnetic domains. A local exchange bias was observed even in not explicitly field-annealed samples, caused by interfacial uncompensated magnetic spins. These experiments provide highly desired information on the relative orientation of electron spins at the interface between ferromagnets and antiferromagnets.

Atomically engineered magnetic thin-film structures are used in a great variety of devices, such as magnetic data storage media in computer hard drives, magnetic sensors or in future applications such as non-volatile magnetic random access memory (MRAM) [1]. Antiferromagnetic layers are an important and scientifically challenging component in these devices. The antiferromagnet magnetically pins or “exchange biases” the magnetization of a ferromagnetic layer to serve as a magnetic reference, resulting in a uniaxial magnetic anisotropy [2]. Due to its compensated magnetic structure antiferromagnets are insensitive to applied magnetic fields. On the other hand, the lack of a macroscopic moment impedes the investigation of the magnetic properties of antiferromagnetic thin films, explaining the ongoing controversies on the origin of exchange bias. Knowledge of the interfacial magnetic structure in the ferromagnet and the antiferromagnet is essential for a correct microscopic description, in particular for an evaluation of the validity of recently developed models, which describe exchange bias either as an effect of spin canting in the antiferromagnet (spin-flop) or as a result of uncompensated surface spins, originating from surface imperfections such as steps, grain boundaries or domain walls [2-4]. The nm-μm sized domains in antiferromagnetic (AFM) thin films require the application of high-resolution techniques for domain imaging, because of the limited sensitivity and spatial resolution of traditional techniques such as neutron diffraction, x-ray diffraction or optical microscopy.

The photoelectron emission microscope PEEM2 located at beam line 7.3.1.1 of the Advanced Light Source (ALS) offers high spatial resolution (typically down to 50 nm) in conjunction with magnetic contrast for the investigation of ferro- and antiferromagnetic thin film structures [5]. Photoelectrons emitted from the sample, which is illuminated by monochromatic x-rays from an ALS bending magnet, are imaged by an electron microscope. Magnetic contrast arises from the dependence of the absorption coefficient and thereby the intensity of electron emission on the relative orientation of the x-ray polarization, which can be varied from linear to left and right circular, and the orientation of the magnetic axis. This effect is called x-ray magnetic dichroism. X-ray magnetic linear dichroism (XMLD) contrast, using linearly polarized x-rays, has been applied in the investigation of two thin film systems, $LaFeO_3$ and NiO.

$LaFeO_3$ is an insulating antiferromagnet, which is of interest because properties like its Néel temperature can be easily adapted to the demands of an application by e.g. Sr doping [6]. Thin $LaFeO_3$ films (40 nm) were grown by oxide molecular beam epitaxy (MBE) by means of a block-by-block growth method on a $SrTiO_3$ single crystal substrate. This method has been shown to yield high-quality epitaxial films [7]. In Fig. 1 PEEM images using linearly polarized x-rays acquired at two photon energies (A = 721.5 eV and B = 723.2 eV) are shown. The electric field vector lies in the sample plane. At these energies the absorption spectrum shows a strong magnetic linear dichroism at the Fe L_3 and L_2 resonances (Fig. 1 bottom). Dividing the images taken at the L_2 resonance produces the XMLD image. Different brightness in this image corresponds to different angles between the antiferromagnetic axis, which is defined by the direction of the magnetic moments in the antiferromagnet and the x-ray polarization vector. The angular dependence of the XMLD intensity is given by $3\cos^2\theta - 1$, with θ the angle between x-ray polarization and the AFM axis [8]. The strong magnetic contrast in the XMLD image arises from magnetic domains with an in-plane projection of the AFM axis parallel (white) and perpendicular (black) to the horizontal x-ray polarization. Temperature dependent experiments have confirmed the magnetic origin of the image contrast, which vanishes approaching the Néel temperature of the antiferromagnet [8]. Furthermore, on Co/$LaFeO_3$ bi-layers, which have been grown by MBE on $SrTiO_3$(100), magnetic exchange coupling across the ferromagnet-antiferromagnet interface causes a clear alignment of the ferromagnetic and the antiferromagnetic domain structures (not shown) [9]. These results demonstrate that photoelectron emission microscopy (PEEM) experiments using synchrotron radiation are capable of determining the magnetic domain structure in thin antiferromagnetic films, within the limits of the spatial resolution of these instruments.

The next example, thin NiO films grown by MBE on a MgO(100) substrate, will demonstrate that although the spatial resolution of the PEEM2 instrument is not sufficient to resolve single antiferromagnetic domains in this sample, useful information can be obtained by magnetic x-ray spectro-microcopy [10]. NiO thin films are of technological importance as antiferromagnetic layers in exchange biasing applications and can be considered a model system to study. Figure 2 shows a magnetic XMLD image of the surface of a 80 nm thick NiO(100) film. The XMLD image was obtained by dividing images acquired at the Ni L_2 resonance at A = 870.3 eV and B = 871.5 eV. This procedure eliminates topographical contrast and enhances the antiferromagnetic contrast. The image exhibits straight bright lines, between 400 nm and 2000 nm wide on a darker background. Similar structures were found at other locations and on other samples with different NiO thickness in the 10-80nm range. The temperature dependence of the magnetic dichroism effect is apparent in local absorption spectra shown in Fig. 3 (top). One series of spectra was acquired in one of the bright stripes, the second in the surrounding darker area. Black arrows mark the photon energy at which PEEM images were obtained. The relatively higher intensity at A compared to B (at 295 K) signifies a predominantly out-of-plane orientation of the antiferromagnetic axis in both regions at room temperature. The effect is more pronounced in the dark area. However, since rotation of the sample around the surface normal does not reverse this effect (not shown), we deduce that the smaller intensity ratio in the lines is not caused by an in-plane rotation of the antiferromagnetic axis (which would break the rotational isotropy) but results from a reduced out-of-plane moment averaged over the spatial resolution of the microscope. Approaching the Néel temperature of NiO (523 K), the intensity ratio of peak A and B approaches the same value in both areas (Fig. 3, bottom), a value which is characteristic for paramagnetic NiO above the Néel temperature, demonstrating the magnetic origin of the image contrast. An analysis of the image intensities at energy A and B within a white line compared with the intensities in the surrounding area as function of temperature furthermore (not shown) allows a local determination of the Néel temperature, which appears to be reduced in the white lines which also exhibit – as explained above – a reduced out-of-plane magnetic moment. Similar line-like structures, consisting of raised bars, arranged in a crisscross pattern were observed in atomic force microscopy images (not shown). Atomic force microscopy is only sensitive to the topography of the sample.

These results convincingly show the usefulness of x-ray spectro-microscopy and PEEM for the determination of the magnetic structure of antiferromagnetic surface and thin films. These techniques in combination with ferromagnetic imaging exploiting x-ray magnetic circular dichroism (XMCD) [11] provide valuable information for an improved understanding of the exchange bias phenomenon. Our inability to resolve the domain pattern in NiO highlights the need for improved instruments with higher spatial resolution for the investigation of polycrystalline films and technologically important materials.

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Fig.1: Antiferromagnetic domain structure in $LaFeO_3$ thin films. The PEEM images were acquired at A = 721.5 eV and B = 723.2 eV at the Fe L_2 edge using linearly polarized x-rays. Areas of different brightness in the divided XMLD image B/A correspond to antiferromagnetic domains with different projection of the antiferromagnetic axis on the x-ray polarization vector E. The measurement geometry is shown top right. Local absorption spectra acquired in single domains show the origin of the image contrast.

Fig.2: XMLD image of NiO(100) thin film obtained by dividing two PEEM images acquired at the Ni L_2 edge. The white lines are regions of reduced out-of-plane magnetization.

Fig. 3: Comparison of local absorption spectra acquired in a white line and in the surrounding dark matrix as function of temperature. The photon energies at which the PEEM images were acquired are marked by arrows (A,B). The ratio of the peak intensities A/B in bright and dark areas is shown below for a subset of temperatures. At low temperature (295 K), the peak ratio A/B>1.05 indicates a predominantly out-of-plane magnetization. Approaching the Néel temperature the ratio approaches the peak ratio for non-magnetic NiO, which is near 1.

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Fig. 1

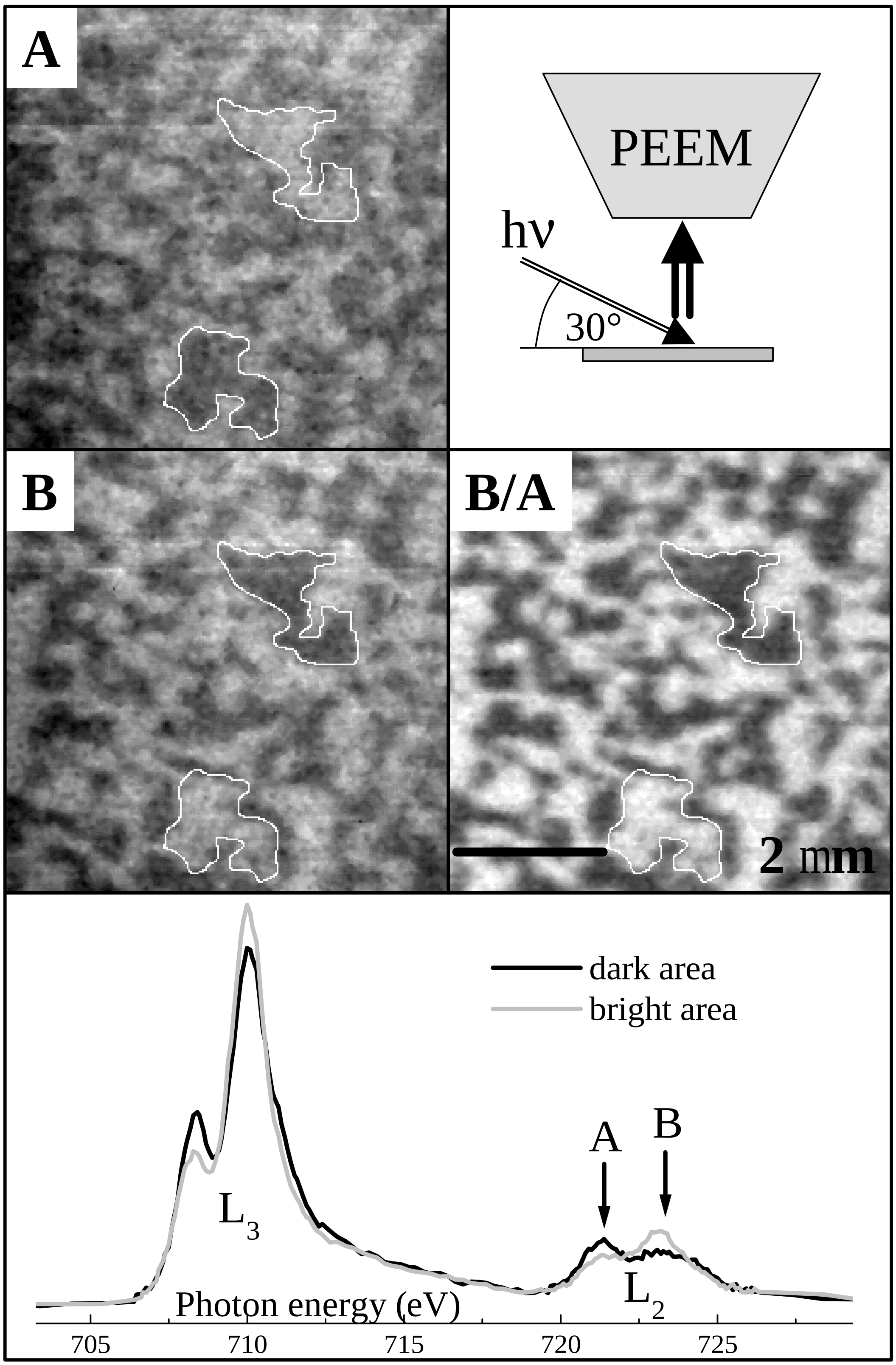

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Fig. 2

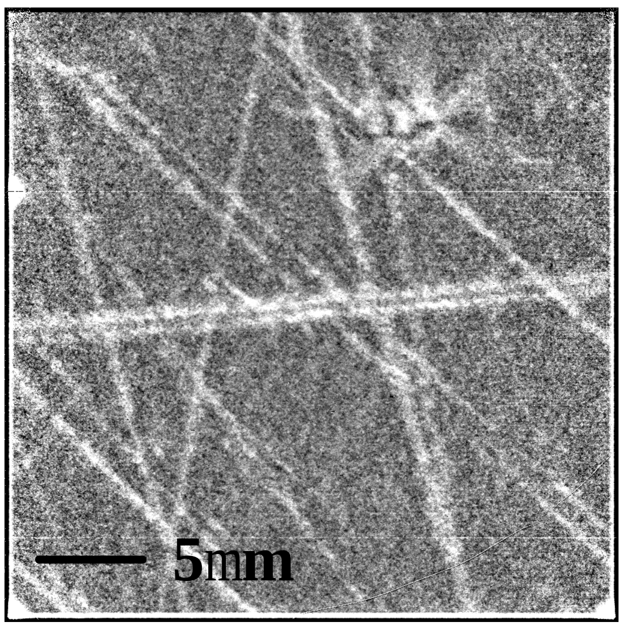

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Fig. 3