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Magneto-dichroic effects in energy- and angle-resolved photoemission: contrast mechanisms for the elementally sensitive imaging of magnetic domains

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Abstract. A number of magneto-dichroic effects has recently been reported in energy- and angle-resolved core-level photoemission with both circularly and linearly polarized light. These various dichroisms can be employed to obtain magnetic sensitivity in photoelectron microscopy. A combination of linear (MLDAD) and circular magnetic dichroism in angle-resolved photoemission (MCDAD) may provide complementary information about the domain structure.

The recent discovery of magneto-dichroic effects in x-ray absorption [1,2] and photoemission [3] led to the advent of an exciting new field in magnetism. This development was strongly supported by the increased availability of circularly polarized synchrotron radiation, which is a necessary requirement for most of the present investigations. The different forms of dichroism that occur in the emitted electrons turn out to be particularly useful in surface and thin-film magnetism, as they combine magnetic and surface sensitivity. The current popularity of magnetic dichroism may be easily understood when considering it as an 'internal spin detector'. The magnetic information is extracted from a simple intensity measurement, instead of involving a time-consuming spin-polarization analysis of the emitted electrons. By studying magnetic dichroism from core-level electrons with well defined binding energies one additionally gains chemical selectivity.

Besides its undisputed usefulness in spectroscopic studies of magnetic phenomena, magnetic dichroism with circularly polarized light may be employed to obtain spatially resolved magnetic information from surfaces by means of direct imagine techniques. Based on reports of a strong MCD signal in the total photoyield at the Ni $L_{2,3}$ edges [4], the first successful imaging of written domains in a magnetic storage material was performed with an immersion-lens photoelectron microscope [5]. The strong electrostatic fields used in this type of instrument effectively integrate the photocurrent over energy and angle. The magnetic contrast arises solely from spatial variations in the total photoyield when irradiating the sample with circularly polarized light of opposite helicities. Since the total photoyield is a measure of the x-ray absorption cross section, the dichroic signal A varies as $A \sim \sigma \cdot M$.

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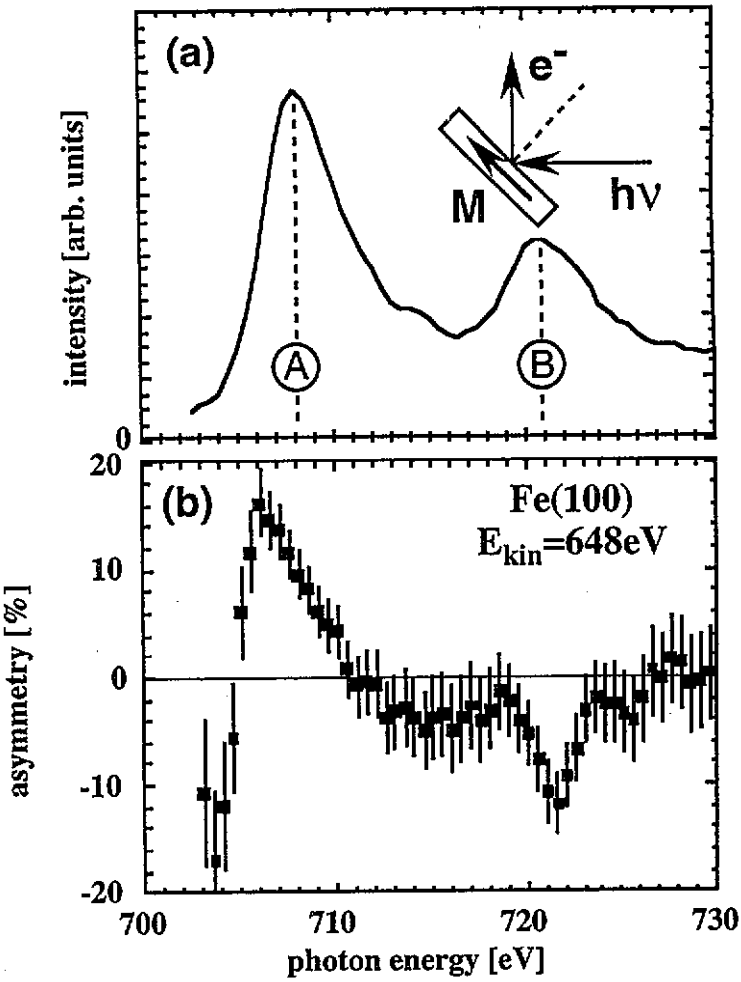

Figure 1. Magnetic circular dichroism in angle-resolved constant-final-state spectroscopy using Fe LMV Auger electrons of 648 eV kinetic energy: (*a*) total intensity and (*b*) intensity asymmetry. Inset: experimental geometry.

A large dichroic signal requires a maximum projection of the photon spin σ onto the domain magnetization M and thus limits the possible experimental geometries.

The above approach suffers from a lack of energy and angular resolution, as other forms of dichroism cannot be exploited for magnetic imaging. Analysing the emitted electrons with respect to their momentum vector and kinetic energy, however, gives access to a complex variety of dichroic phenomena. Particularly in single crystals, the photoelectron wave vector k and the crystalline axes play important roles in addition to σ and M [6]. This leads to magneto-dichroic effects in the angular distribution of photoelectrons with both circularly (MCDAD) and linearly polarized light (MLDAD) [7]. Each of these effects involves a specific experimental geometry. A direct-imaging experiment may utilize these characteristics to obtain complementary information about the local orientation of the magnetization vector M. In order to take advantage of these magnetic dichroisms, an energy-resolving technique is needed. Our approach employed a VG Escascope, which consists of a hemispherical

capacitor as energy analyser and a specially adapted electrostatic lens system [8]. In contrast to an immersion lens this arrangement allows measurements even at high tilt angles and retains a certain angular resolution, since no accelerating field are applied between sample and lens system. The imaging experiments were performed on Fe(100) whiskers, which are known to exhibit well defined domain patterns. Spectroscopic studies were carried out either on Fe single crystals mounted on a U-shaped soft-iron yoke equipped with a magnetizing coil, or on Fe whiskers using the 'small-spot' mode of the Escascope. Circularly polarized soft x-rays were obtained from the SX-700-3 monochromator at the German storage ring BESSY [9].

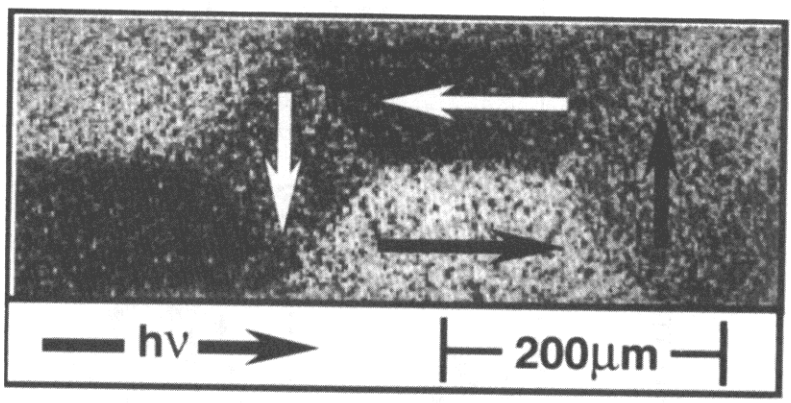

Figure 2. Magnetic domain pattern from an Fe(100) whisker obtained with Fe LMV Auger electrons (contrast mechanism shown in figure 1) by means of energy-resolving electron microscopy. Arrows indicate the magnetization direction in each domain (for details see text).

First, we will illustrate the advantage of *energy resolution* in a direct-imaging experiment. Auger electrons, which appear as secondary electrons in a photoemission process, show a strong magnetic circular dichroism if the excitation energy is tuned close to the associated photoemission threshold [10]. This behaviour is shown in figure 1 for the Fe LMV Auger electrons. The light was incident at 65° with respect to the surface normal. The peak intensity at the $2p_{3/2}$ (A) and $2p_{1/2}$ (B) photoexcitation thresholds for a given magnetization direction depends strongly on the helicity. This gives rise to a pronounced magneto-dichroic signal, which is expressed as the intensity asymmetry $A = (I^+ - I^-)/(I^+ + I^-)$ of spectra obtained at opposite light helicities. In order to separate the magnetic contrast in a photoelectron image from other contrast mechanisms such as topography or chemistry, two images with reversed magnetic information (usually taken at opposite helicities) must be subtracted. Figure 1 suggests a variation of this procedure as the value of A is about the same absolute size and of opposite sign for the $j = \frac{3}{2}$ and $j = \frac{1}{2}$ levels. The grey-scale domain image from an Fe(100) whisker obtained by subtracting images acquired at 708 eV and 721 eV photon energy shows a region where domains form a flux-closure pattern (figure 2). The two domains in the centre of the image are separated by a straight 180° wall and are oriented parallel (white) and antiparallel (black) to the photon spin σ. The magnetization in the diamond-shaped domains on either side of this configuration points perpendicular to σ. They appear therefore in the same medium-grey level, although their magnetization vectors are mutually opposite. The experiment cannot distinguish between these domains unless the sample is rotated so that a sizeable projection

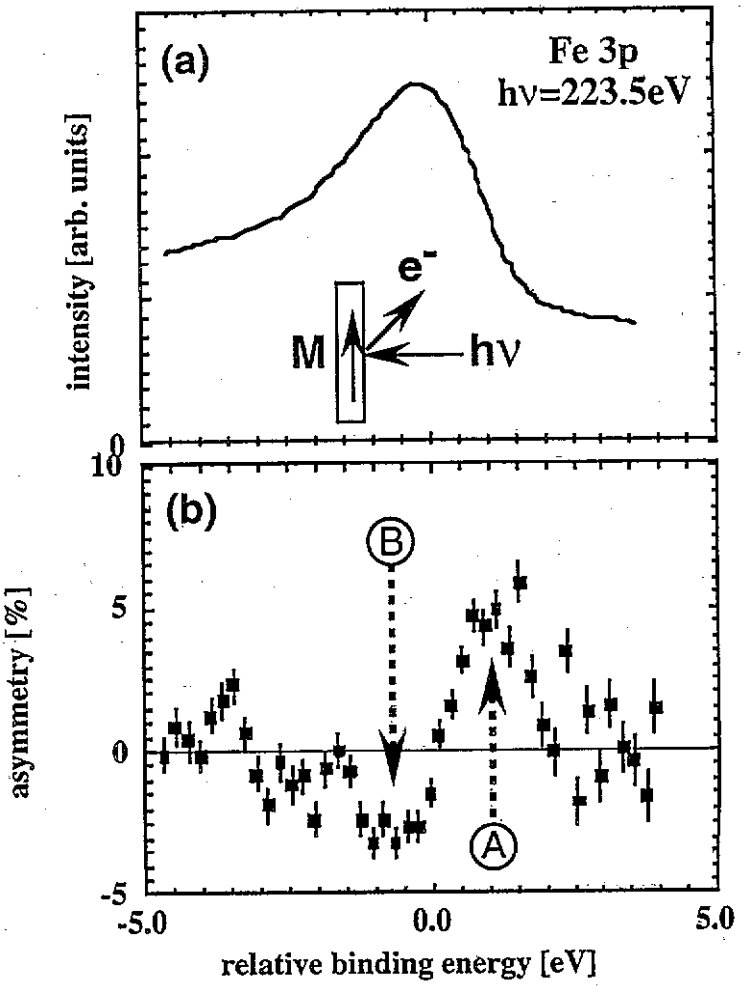

Figure 3. Magnetic circular dichroism in angle-resolved photoemission (MCDAD) from the Fe 3p levels: top panel, total intensity, and bottom panel, intensity asymmetry. Inset: experimental geometry.

of their magnetization points along σ. This angular dependence is due to the fact that Auger electron emission incorporates contributions from photoexcitations into all possible final states. This situation corresponds to an internal angular integration with respect to the final-state selection effects discussed in the following paragraph, and leads to an angular variation of the dichroic signal as $A \sim \sigma \cdot M$. The energy resolution offers a better signal-to-noise ratio in the magnetic contrast as compared to a total-yield experiment. It furthermore introduces an additional depth selectivity through the choice of widely different electron energies, for example, from different Auger transitions.

The above approach can be further refined by additionally introducing *angular resolution* into the experiment and imaging with direct photoelectrons rather than secondary electrons. The dichroic asymmetry in the energy- and angle-resolved photocurrent exhibits completely different behaviour, since the photoelectron wave vector k now becomes explicitly involved. It has been shown for MCDAD in 2p photoemission (assuming predominantly p→d

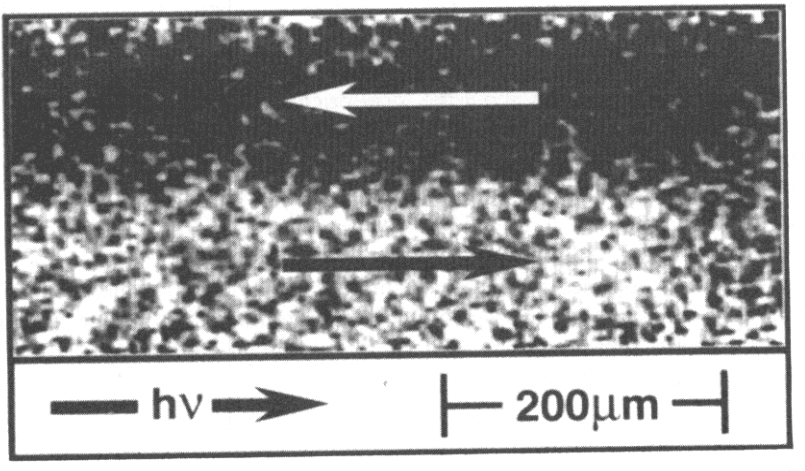

Figure 4. Magnetic domain pattern from an Fe(100) whisker obtained by means of MCDAD with Fe 3p photoelectrons using energy- and angle-resolved photoelectron microscopy (experimental geometry similar to figure 1; angle of incidence 55°). Arrows indicate the domain magnetization.

transitions) that the asymmetry varies as $A \sim (\boldsymbol{\sigma} \cdot \boldsymbol{M}) - (\boldsymbol{\sigma} \cdot \boldsymbol{k})(\boldsymbol{k} \cdot \boldsymbol{M})$ [6]. As an important consequence, systems magnetized in plane may exhibit a strong magneto-dichroic signal even for normally incident light, i.e. $\boldsymbol{\sigma}_{\perp} \boldsymbol{M}$. This is in contrast to the grazing-incidence geometry required by the same systems if angular integration is involved. Results for Fe 3p photoemission are displayed in figure 3. The dichroic asymmetry (figure 3(*b*)) is characterized by a single plus/minus feature. Again this form of asymmetry suggests that the magnetic contrast can be separated by subtracting two images taken at different kinetic energies corresponding to the extremal values of A (marked by A and B). A domain image acquired with this contrast mechanism is reproduced in figure 4 (except for the angle of incidence of 55° the experimental geometry is the same as given in the inset in figure 1). It was obtained from the same whisker as imaged in figure 2 but shows a different area where only a single straight 180° wall is present. The two oppositely magnetized domains are easily distinguishable by their contrast. We note that the signal-to-noise ratio is not as good as that in figure 2. Two mechanisms are held responsible for this. First, due to our present set-up, the angle of incidence was only 55° (instead of the preferred 90°) with respect to the surface normal. Second, the angular acceptance of the Escascope in the mode used is estimated to be about ±15°. Both effects reduce the intensity asymmetry and impair the signal-to-noise ratio. Still, the result shows very clearly that the limited angular resolution of the instrument is sufficient to generate a magnetic contrast on the basis of MCDAD effects.

Both contrast mechanisms discussed above have in common the fact that they require circularly polarized light. Furthermore, they are only sensitive to magnetization directions within the reaction plane spanned by $\boldsymbol{\sigma}$ and $\boldsymbol{k}$. In order to obtain a complete domain pattern of the surface under investigation, the sample must therefore be rotated with respect to this plane. A more elegant solution to this problem, however, is indicated by a recent work, which reports magneto-dichroic effects with linearly polarized light [11]. These studies revealed a non-zero magnetic linear dichroism in angular-resolved photoemission (MLDAD) when the magnetization is *perpendicular* to the direction of incidence of p-polarized light and thus the reaction plane. The asymmetries observed for Fe 3p photoemission under these conditions were of the order of 5% and are thus comparable to the valued observed with

MCDAD (figure 1). Linearly polarized synchrotron radiation is readily available even from bending magnet sections dedicated to the use of the circularly polarized light. By combining both MCDAD and MLDAD one may fully exploit the complementary angular dependences and extract the full information about the local orientation of the magnetization.

In summary, we have successfully employed magnetodichroic effects in the angular distribution of emitted photoelectrons to image magnetic domains. In order to obtain complete information about the magnetic microstructure at the surface we suggest future experiments based on a combination of MCDAD and MLDAD.

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