New Magnetic Linear Dichroism in Total Photoelectron Yield for Magnetic Domain Imaging

F. U. Hillebrecht, T. Kinoshita,* D. Spanke, J. Dresselhaus, Ch. Roth, H. B. Rose, and E. Kisker
Institut für Angewandte Physik, Heinrich-Heine-Universität Düsseldorf, D-40225 Düsseldorf, Germany
(Received 8 May 1995)

We report a dependence of the photoabsorption cross section as measured by total electron yield at the Fe and Co $3p$ thresholds on the *sign* of the magnetization for p-polarized light at oblique incidence. Peak-to-peak asymmetries up to 4% are observed. The asymmetry increases towards grazing incidence, where the total sample current is smallest. The transverse magneto-optic Kerr effect measured simultaneously shows a peak-to-peak asymmetry of up to 23% at the Co $3p$ threshold. The dichroism is used to image magnetic domains on an Fe(100) surface in a photoelectron emission microscope.

PACS numbers: 78.20.Ls, 75.60.Ch, 78.70.Dm

Spectroscopy of the core levels of the ferromagnetic transition elements has shown in recent years to be surprisingly rich in features caused by the Coulomb and exchange interactions between the core hole and the spin-polarized valence electrons. X-ray absorption experiments (XAS) with linearly polarized light [1] show a change of the spectra when the polarization vector is either parallel or perpendicular to the magnetization axis, according to the selection rules $\Delta m = 0$ or $\Delta m = \pm 1$, respectively. Magnetic circular dichroism (MCD) in x-ray absorption [2,3] can be understood as a spin-dependent excitation of core electrons into empty states above the Fermi level, which—because of the ferromagnetism—are spin polarized [4]. The spin dependence in the excitation arises from coupling between light helicity and the orbital momentum of the excited electron, which is in turn coupled to the spin of the photoexcited electron by spin-orbit interaction. Sum rules, which allow one to extract quantitatively spin and orbital moments, make MCD in absorption a very attractive technique [5]. By measuring the photoabsorption cross section via the total photoelectron yield of the sample, it is even possible to obtain spatially resolved and chemically specific magnetic information [6].

In photoemission, even though the final state of the photoelectron far above the Fermi level has only negligible exchange and spin-orbit interactions, related effects can also be observed. It is well known that core level photoelectrons from ferromagnets in general carry a spin polarization due to exchange [7]. Apart from magnetic circular and "conventional" (as described above for XAS) linear dichroism, a new type of magnetic linear dichroism was recently reported for photoemission appearing on magnetization *reversal* [8]. This effect requires that the vectors of magnetization, electric radiation field, and electron emission form a chiral system with the angle between light polarization and electron emission different from 90°, and occurs only in angle-resolved experiments. In photoabsorption, the angular acceptance of the excited electrons is not restricted, so that it is, in principle, an angle-integrating experiment. However, Kao and co-workers [9] reported in 1990 for photon energies near the Fe $2p$ excitation thresholds a change of the reflectivity for obliquely incident p-polarized light when the magnetization is switched between the two directions normal on the plane of incidence. This experiment is the soft x-ray analog to the transverse magneto-optic Kerr effect (T-MOKE) known for the visible spectral region [10]. Kao *et al.* report a resonant enhancement of the T-MOKE signal at the Fe $2p$ edges, with a maximum asymmetry of 13% for 17.8° incidence angle. In their pioneering analysis of magneto-optical effects at the Ni $3p$ absorption threshold, Erskine and Stern [11] also noticed that such an effect is to be expected, and may have a relative magnitude of the order of 10%. If there are no nonlinear effects the absorbed intensity (A) should combine with the reflected one (R) so that $R + A = 1$ (incident intensity normalized to 1). Consequently, there should be a dichroism in this geometry also in absorption at a core level threshold, which exhibits a transverse magneto-optic Kerr effect.

We report here on the observation of this type of magnetic *linear* dichroism in the total photocurrent for Fe and Co in the vicinity of the $3p$ excitation threshold, and its use for the imaging of magnetic domains at the surface of an Fe(100) single crystal. The dichroism shows up with p-polarized light as a dependence of the photocurrent on the *sign* of magnetization in transverse geometry for photon energies near the $3p$ core level thresholds of Fe and Co (53 and 58 eV, respectively). For Co $3p$, we simultaneously observe a very large magneto-optic Kerr effect, analogous to the result of Kao *et al.* [9] for the Fe $2p$ level. Although most of our results are for ultrathin epitaxial Fe and Co films grown on W(110) and Cu(100), respectively, the dichroism is not a thin film effect as we obtain the same dichroism spectrum from magnetic domain imaging performed on an Fe single crystal surface.

The experiments were performed at the crossed undulator beamline U2 at BESSY [12] and the planar undulator BW3/SX 700-beamline at HASYLAB [13]. The crossed undulator offers both linear light polarizations (p and s) as well as partial circular polarization, the helicity of which can be switched via a magnet structure

 0031-9007/95/75(11)/2224(4)$06.00

situated between the two planar undulators. The planar undulator at HASYLAB provides linearly p-polarized light. For total yield measurements the sample was grounded via a picoamperemeter, while the reflected light was measured via the current released from a gold diode. The samples were films of Co on Cu(100) and Fe on W(110) grown *in situ* in UHV. The films grow epitaxially in bcc (Fe) and fcc (Co) structure. Film thicknesses were 10–30 monolayers for Co, and about 50 monolayers for Fe, yielding easy axes along the [110] and [100] in-plane directions of Co/Cu and Fe/W, respectively. The samples were magnetized by current pulses through magnetization coils mounted nearby; measurements were carried out in remanence. Figure 1 shows the geometry for spectroscopic and imaging experiments. Domain images were obtained at the BESSY beamline from a sputtered Fe(100) single crystal sample using an electrostatic photoemission microscope (PEEM 150 by Staib Instruments) originally described by Engel *et al.* [14] in which the secondary electron cascade is imaged to the channel plate without explicit energy filtering. For microscopy the incidence angle of the light (θ) was 75°. As the secondary electron cascade peaks at low kinetic energy, the angular acceptance is so large that the instrument effectively measures the total photocurrent in a spatially resolved way. Kerr microscopy of the Fe crystal showed irregular domains. To obtain magnetic contrast in the PEEM, images obtained with different photon energies in the vicinity of the $3p$ threshold were subtracted from each other.

Figure 2(a) shows the spectral dependence of the total current excited by s- and p-polarized light in the vicinity of the Fe $3p$ threshold for $\theta = 75°$. The shape of the photocurrent curve differs from the shape observed at normal incidence, where we obtain a line shape similar to the so-called white line [15], i.e., a strong sharp resonance in the absorption cross section due to dipole transitions into the empty portion of the d band [16]. For p-polarized light, the shape of the total yield curve is affected when the magnetization is reversed; i.e., there is a magnetic dichroism. Figure 2(b) shows the corresponding asymmetries, defined as $(I^{\uparrow} - I^{\downarrow})/(I^{\uparrow} + I^{\downarrow})$, where $I^{\uparrow}$ and $I^{\downarrow}$ are the current yields with the sample magnetized in the two transverse directions. With s-polarized light, the total yield curve does not depend on the direction of the magnetization. Figure 3 shows the corresponding data for the Co surface (with p-polarized light only). The peak-to-peak asymmetry in the total yield was ≈2.5% for Fe and Co. Figure 4(a) shows the optical reflectivity of the Co surface for the same geometry with p-polarized light, obtained quasisimultaneously with the total yield. The reflectivity exhibits a broad, approximately 20 eV wide dip, centered slightly (1 eV) above the energy where the Co $3p$ photoabsorption curve (Fig. 3) sets in. Figure 4 shows that also the reflectivity is strongly affected when the magnetization is reversed. The effect of magnetization reversal is much stronger in the reflectivity than in the total yield measurement, yielding a peak-to-peak asymmetry of 22.5%. The reflectivity results are qualitatively similar to those of Kao *et al.* [9] for the Fe $2p$ line.

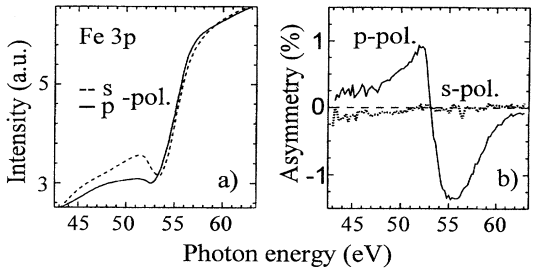

FIG. 2. (a) Total photoyield near the Fe $3p$ edge measured with s- and p-polarized light incident at 75° to the surface normal as a function of photon energy. (b) The associated asymmetries. The sample magnetization was reversed between the two directions normal on the plane of light incidence.

In a macroscopic description, T-MOKE stems from the complex off-diagonal element ε_{12} of the dielectric tensor [10,11]. This quantity is linear in M and thus causes the dependence of reflected light intensity on the sign of magnetization in the transverse geometry. In the visible part of the light spectrum this effect is less than 1%. The large asymmetry in the core level energy region is related to the absorption (ε_{12}-) resonance caused by the presence of the core hole. Since the asymmetries in absorption and reflection are related to each other, one may discuss both these effects within the dielectric theory of ε_{12}. On the basis of the optical sum rule $R + A = 1$ the asymmetries in absorption ($\Delta A/A$) and reflectivity ($\Delta R/R$), with A and R being the mean absorption or reflectivity, respectively,

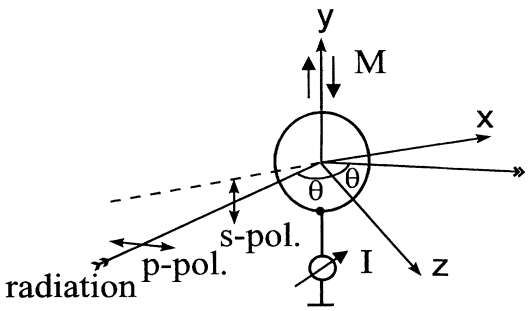

FIG. 1. Experimental geometry.

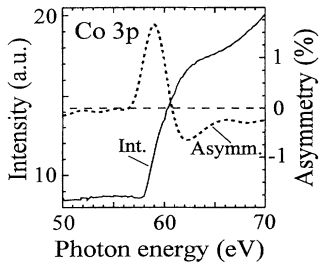

FIG. 3. Same as Fig. 2, but for the Co $3p$ core level (p-polarized light only).

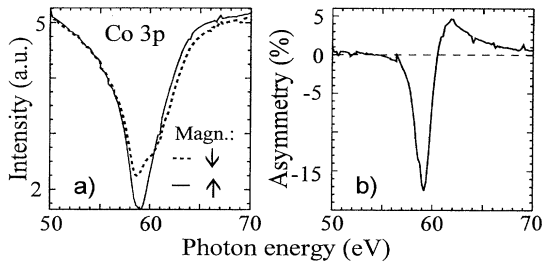

FIG. 4. Specular optical reflectivities (a) and associated asymmetry (b) for Co under identical conditions as in Fig. 3.

should be related as $\Delta A/A = -R/(1 - R)\Delta R/R$. This shows that for strong absorption, i.e., small reflectivity, the asymmetry in the reflectivity can be much larger than in absorption. Similar trends have been observed with circularly polarized light [17].

Figure 5 shows the dependence of the asymmetries on the angle of light incidence for photoabsorption and for Co also for reflection. In photoabsorption, the asymmetry increases towards bigger angles, i.e., towards more grazing incidence, while the average yield (not shown) decreases. In contrast to that, the asymmetry in reflection (T-MOKE) increases with decreasing angle of incidence. The dependence of the intensity difference in transverse MOKE [10] on the angel of incidence θ is given by a product of the (modified) Fresnel coefficient for p-polarized light and a term $\varepsilon_{12} \sin 2\theta/(n_0^2 \cos^2\theta - 1)$. On the basis of the optical sum rule, we expect the differences ΔR and ΔA to be of equal size, with maxima occurring at the same angle of incidence. For the refractive index n_0 in the range 0.95 to 1.05, the largest difference should occur for $\theta > 75°$. The maxima of the asymmetries $\Delta A/A$ and $\Delta R/R$, however, may occur at different angles, due to the angle dependences of A and R. Specifically, we obtain reflectivities of about 0.5 and 0.1 for 85° and 75° incidence angles, inserting

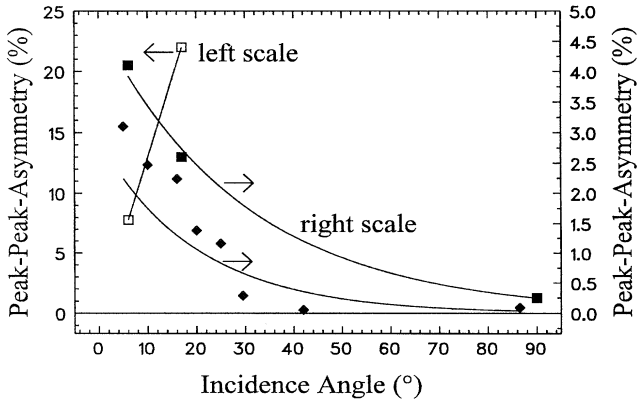

FIG. 5. Angle dependence of the linear magnetic dichroism in terms of the peak-to-peak asymmetry in the total yield (sample current) for Fe (diamonds) and Co (squares) and of the specular optical reflectivity (open squares) for Co.

standard optical constants for the threshold region [18] into the Fresnel equation. Consequently, for $\theta = 85°$, the asymmetries for absorption and reflection should be of similar magnitude, $\Delta A/A \cong -\Delta R/R$, while for 75° one obtains a ratio of 10:1 between them. This is in good agreement with the experimental data shown in Fig. 5. In general, for R approaching 1 towards grazing incidence, the maximum asymmetry in absorption is shifted towards more grazing incidence from the maximum asymmetry in reflectivity due to the angle dependence of the factor $R/(1 - R)$. This illustrates why $\Delta A/A$ does not show a maximum in the range accessible in our experiment. Also, the angular dependence of the reflectivity asymmetry does not show a maximum due to the limited angle range investigated. From the foregoing it appears that the magnetic linear dichroism in the total yield in transverse geometry is a surface effect, which makes it particularly suitable for the study of surface magnetism. The theoretical treatment of x-ray resonant exchange scattering by Hannon *et al.* [19] leads to an angle dependence of the intensity asymmetry according to $\varepsilon_{12} \sin^2 2\theta/(B \cos^2 2\theta + C \sin^2 2\theta)$ [9,19]. Also in this case one expects an initial increase of the effect as one moves from grazing towards more normal incidence, and the dichroism in absorption as well as in reflection has to disappear as $\theta = 0°$ or 90° is approached, in agreement with our experimental result.

We have used the dichroism in the total photocurrent for imaging magnetic domains at the surface of an unmagnetized Fe(100) single crystal. Images were taken with p-polarized light, with photon energies between 48 and 62 eV. In domain imaging using linearly polarized light, there is no equivalent of reversing the helicity. Therefore, we exploit the dependence of the asymmetry on the photon energy to obtain two images that contain the domain information in different magnitude. The largest magnetization-induced difference of the total yield is obtained for 52 and 55 eV photon energies [see Fig. 2(b)]. Figure 6 shows an image constructed by subtraction of images taken with p-polarized light of 52 and 55 eV after scaling to the same intensity. The field of view has 340 μm diameter. Data acquisition time for this image was about 50 s. While the original images are similar to each other, the difference image shows a bright area and a dark area, which do not correspond to structures in the original images. This contrast is attributed to a domain structure. The width of the transition between the two regions corresponds to a spatial resolution of 5 μm in this low magnification mode of the microscope. The asymmetry extracted from a series of images as a function of the photon energy resembles very closely the data shown in Fig. 2(b). This demonstrates that the regions of different brightness visible in Fig. 6 are indeed caused by magnetic domains, with magnetization directions as indicated. Furthermore, the agreement between the asymmetry obtained for the Fe(001) surface and that of the thin film shows that the

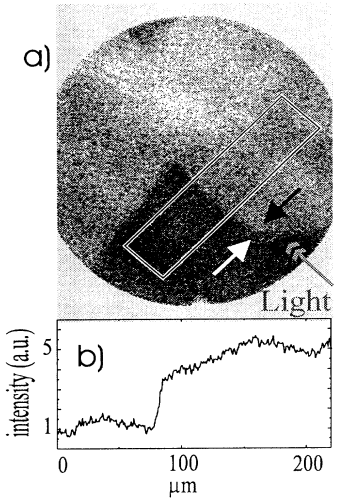

FIG. 6. Magnetic domains of an Fe(100) single crystal, determined by photoemission microscopy excited by linearly p-polarized light incident under 75° to the surface normal. The domain image is a difference of images taken with 52 and 55 eV photon energies.

magnetic dichroism is not related to the finite thickness of the thin film samples.

In summary, we report a new type of magnetic linear dichroism in photoabsorption measured via the total yield in transverse magnetization geometry. It shows up on *reversal* of the magnetization, using p-polarized light. The advantage to be gained by using soft x rays instead of light in the visible region is, as with other soft x-ray techniques, that core level binding energies are element specific, so that magnetic properties can be probed individually for each of the constituents in compound materials. The dichroism offers contrast to be used to image magnetic domains via the total electron yield in a photoemission microscope. The attainable resolution can be higher than with the commonly used magneto-optical effects in the visible or near-visible spectral region due to the shorter wavelength in core level spectroscopy. Also, magneto-optical effects in the visible are usually small, whereas we observe at the Co $3p$ core level binding energy the largest transverse magneto-optic Kerr effect (23% p-p) reported so far. Furthermore, with the large magnetic effects observed in the reflectivity, a soft x-ray optical Kerr microscope seems feasible.

We wish to thank A. Bringer and M. Schäfer, both at IFF, KFA Jülich, for helpful discussions and Kerr microscopy, respectively. We are grateful to G. Reichardt and A. Gaupp of BESSY and to Th. Möller of HASYLAB for their assistance in running the beamlines, as well as U. Rosowski, S. Manderla, and W. Schützek of the Institut für Angewandte Physik for their expert technical help. This work was supported by the Deutsche Forschungsgemeinschaft (SFB 166/G7) and by the Federal Ministry for Research and Technology (BMFT, 05 5PFDAB 3). The photoemission microscope was purchased with funds by the Ministerium für Wissenschaft und Forschung of the state of Nordrhein-Westfalen.

*Humboldt fellow on leave from Institute for Solid State Physics, University of Tokyo, Roppongi, Minato-ku, Tokyo 106, Japan. Present address: UVSOR, Institute for Molecular Science, Okazaki 444, Myodaiji, Japan.

[1] G. van der Laan *et al.,* Phys. Rev. B **34**, 6529 (1986); J. B. Goedkoop *et al.,* Phys. Rev. B **37**, 2086 (1988); R. J. H. Kappert *et al.,* Surf. Sci. Lett. **248**, L245 (1991).

[2] G. Schütz, W. Wagner, W. Wilhelm, P. Kienle, R. Zeller, R. Frahm, and G. Materlik, Phys. Rev. Lett. **58**, 737 (1987); G. Schütz, M. Külle, and H. Ebert, Phys. Scr. **T49**, 302 (1993).

[3] C. T. Chen, F. Sette, Y. Ma, and S. Modesti, Phys. Rev. B **42**, 7262 (1990); C. T. Chen, N. V. Smith, and F. Sette, Phys. Rev. B **43**, 6785 (1991).

[4] H. Ebert, P. Strange, and B. L. Gyorffy, Z. Phys. B **73**, 77 (1988).

[5] B. T. Thole *et al.,* Phys. Rev. Lett. **68**, 1943 (1992); P. Carra *et al.,* Phys. Rev. Lett. **70**, 694 (1993).

[6] J. Stöhr *et al.,* Science **259**, 658 (1993).

[7] C. Carbone and E. Kisker, Solid State Commun. **65**, 1107 (1988).

[8] Ch. Roth, F. U. Hillebrecht, H. B. Rose, and E. Kisker, Phys. Rev. Lett. **70**, 3479 (1993).

[9] C. Kao, J. B. Hastings, E. D. Johnson, D. P. Siddons, G. C. Smith, and G. A. Prinz, Phys. Rev. Lett. **65**, 373 (1990).

[10] M. J. Freiser, IEEE Trans. Magn. **4**, 152 (1968).

[11] J. L. Erskine and E. A. Stern, Phys. Rev. B **12**, 5016 (1975).

[12] J. Bahrdt, A. Gaupp, W. Gudat, M. Mast, K. Molter, W. B. Peatman, M. Scheer, Th. Schroeter, and Ch. Wang, Rev. Sci. Instrum. **63**, 339 (1992).

[13] C. U. S. Larsson, A. Beutler, F. Federmann, A. Rieck, S. Verbin, and T. Möller (unpublished); T. Möller, Synchrotron Radiat. News **6**, 16 (1993).

[14] W. Engel, M. E. Kordesch, H. H. Rotermund, S. Kubala, and A. von Oertzen, Ultramicroscopy **36**, 148 (1991).

[15] K.-H. Walker, E. Kisker, C. Carbone, and R. Clauberg, Phys. Rev. B **35**, 1616 (1987).

[16] The total yield is proportional to $d/(d + l\cos\theta)$, where d is the mean free path of the secondary electrons, l is the mean free path of the photons (i.e., inversely proportional to the excitation cross section), and θ the angle of light propagation in the material measured from the surface normal [B. T. Thole *et al.,* Phys. Rev. B **32**, 5107 (1985)]. The absence of the white line indicates that the photon mean free path projected on the surface normal (i.e., $l\cos\theta$) is comparable to the electron mean free path d [see also H. G. Birken, C. Blessing, and C. Kunz, in *Handbook of Optical Constants of Solids* (Ref. [18]), Vol. II, p. 279].

[17] C.-C. Kao *et al.,* Phys. Rev. B **50**, 9599 (1994).

[18] *Handbook of Optical Constants of Solids,* edited by E. D. Palik (Academic Press, Boston, 1991), Vol. II, p. 385 ff; B. L. Henke *et al.,* At. Data. Nucl. Data Tables **27**, 1 (1982).

[19] J. P. Hannon *et al.,* Phys. Rev. Lett. **61**, 1245 (1988).

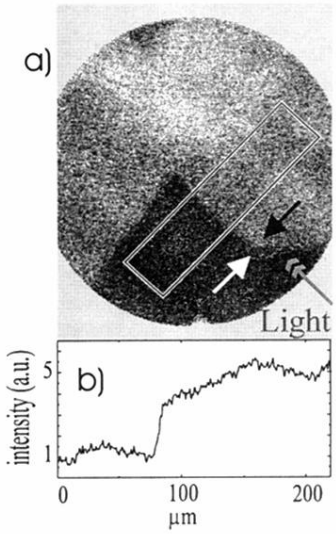

FIG. 6. Magnetic domains of an Fe(100) single crystal, determined by photoemission microscopy excited by linearly *p*-polarized light incident under 75° to the surface normal. The domain image is a difference of images taken with 52 and 55 eV photon energies.