Linear Magnetic Dichroism in Angular Resolved Fe 3*p* Core Level Photoemission

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We have observed a new type of magnetic linear dichroism in angle-resolved, spin-integrated photoemission: For *p*-polarized light under oblique incidence the Fe 3*p* core level peak position and line shape change when the sample magnetization is reversed. Spin-resolved measurements show that the effect is due to spin-orbit interaction in the presence of exchange interaction. The effect can be used for chemically specific diagnostics of magnetic structures.

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The interaction of polarized light with ferromagnetic materials produces a variety of magneto-optical effects, among which the Kerr and Faraday effects are the most important ones. They are observed in optical absorption or reflectance. They are often called magnetic dichroism effects and are used for instance in imaging ferromagnetic domains. The physical basis of the Faraday and Kerr effects is the combined influence of spin-orbit (SO) and exchange interactions on the transitions between the initial and final states involved in the absorption or scattering of the incoming photons [1]. In the 3*d* transition metals, SO is small for the 3*d* valence states probed by visible light, and hence the magneto-optical effects are small in this regime. However, in core levels, SO interaction is larger, and therefore large magnetic dichroism effects can be expected if core levels are involved in the transitions. With the development of synchrotron radiation optics studies of magnetic circular dichroism in photoabsorption have been extended towards the soft-x-ray regime in the pioneering experiments by Schütz *et al.* [2]. In photoemission, the electrons are excited to states far above threshold so that—in contrast to magneto optics or absorption—the final state spin polarization does not play a role. However, the matrix elements by themselves are sufficient to cause a dependence of photoemission spectra on magnetization. Magnetic dichroism in the (spin-integrated) Fe 2*p* core level photoemission spectra when using circularly polarized light have indeed been reported first by Baumgarten *et al.* [3]. In both experiments [2,3] the spectra were found to depend on the relative orientation of photon helicity and sample magnetization vectors. We report here for the first time on a striking dependence of photoemission line shapes on the direction of magnetization when using *linearly* polarized light. This is observed in angle-resolved photoemission from the Fe 3*p* core level. Spin analysis of the 3*p* photoelectrons allows us to uncover the importance of SO interaction in this effect. Our data furthermore are important for the understanding of the Fe 3*p* core level photoemission spectra since they exhibit distinct sublevel states for the first time.

In previous works, the 3*p* spectrum of ferromagnetic Fe has been found to be spin polarized [4,5] despite the fact that the initial state is a closed shell: The line shapes are spin dependent, and there is an overall negative polarization. Until now, these findings have been ascribed to the exchange interaction between the 3*p* core hole created in the emission process and the spin-polarized *d* electrons. Neither the spin-resolved line shapes nor the overall spin polarization are yet understood.

The present experiment was performed using linearly polarized light of 90 eV energy from the new crossed field undulator at BESSY [6]. Here, we took advantage of the fact that the light could be chosen to be *p* or *s* polarized without changing the experimental geometry. A focusing spherical grating monochromator delivered radiation between 20 and 90 eV to the sample, which was a thin Fe(001) film grown epitaxially on Ag(001) or Au(001) substrates. We used a hemispherical photoelectron spectrometer equipped with an Fe(001) very low energy electron diffraction spin polarimeter [7]. The incidence angle of the light was 16° off from grazing and 5° azimuthal with respect to the Fe [100] direction; electrons were collected under normal emission with about 8° full acceptance (see Fig. 1). If SO interaction is present, excitation by *p*-polarized light leads to spin polarization **P** with a quantization axis parallel to **E**×**p** (*p* is the electron

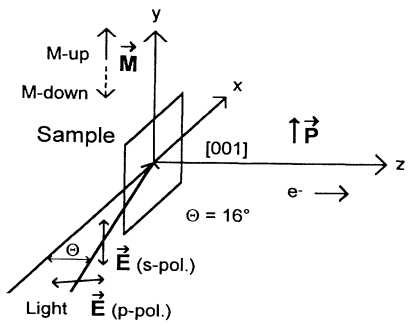

FIG. 1. Experimental geometry: *p*- or *s*-polarized light impinges under 16° onto the Fe(001) surface, nearly parallel to the 10 direction (5° off). Normal emission photoelectrons are collected with 8° full acceptance. The ferromagnetic sample is magnetized remanently parallel ("*M* up") or antiparallel ("*M* down") to the *y* axis.

momentum) [8,9], i.e., in the y direction. The spin is labeled as "up" when it is parallel to the y axis (Fig. 1). The quantization axis for the exchange interaction is the long-range magnetization direction which, in the present experiment, is chosen to be either parallel ("M up") or antiparallel ("M down") to the y direction. In other words, we have chosen a geometry here for which the two quantization axes are collinear. As usual, we refer by majority-spin (minority-spin) emission to those electrons with magnetic moments parallel (antiparallel) to the magnetization direction.

Figure 2(a) shows spin-integrated energy distribution curves (EDCs) of the Fe $3p$ core level obtained for the two directions of the magnetization (M up, M down) with p-polarized light. The Fe $3p$ emission is superimposed on a background of inelastically scattered electrons which increases towards larger binding energy [10]. The integrated intensities of the $3p$ lines above the background are equal. The EDC for M up shows a single asymmetric peak with a smaller reproducible structure on the high-binding-energy side. For M down, the peak is at 0.8 eV higher binding energy. The line is much wider and its appearance suggests that it is composed of three overlapping contributions. This is more clearly seen from our spin-resolved data which are shown below. The energy range in which the spectrum depends on magnetization direction is restricted to 4 eV. Figure 2(b) shows Fe $3p$ EDCs obtained with s-polarized light under otherwise identical conditions from the same sample. These spectra do not depend on the direction of magnetization. Their shape is consistent with those reported previously for the Fe $3p$ level [4,5] with s-polarized light. In order to find the reason for the magnetization dependence of the spin-integrated spectra in Fig. 2(a), we have measured these spectra also spin *resolved*. In Fig. 3, each of the two spin-integrated EDCs of Fig. 2(a) is decomposed into two spin-resolved EDCs, with spin directions up and down referring to the *positive* y direction *independent* of magnetization. With reference to earlier work where SO interaction was not important we have labeled the spin-resolved EDCs also by their spin character with respect to the sample magnetization (majority, minority). For both magnetizations the emission in the minority channel has more intensity than in the majority channel. However, the intensity difference between up and down spins is much larger for M up than for M down.

To understand qualitatively the dependence of the spectra on the direction of the magnetization, we recall that the spin-orbit interaction can cause a spin polarization parallel or antiparallel to $\mathbf{E}\times\mathbf{p}$ when the light is p polarized and incident under an angle $0° < \Theta < 90°$. This was found experimentally in photoemission from the Ar $3p$ and Xe $5p$ core levels [9] and in angle-resolved photoemission from high-Z metals [11]. Theoretical

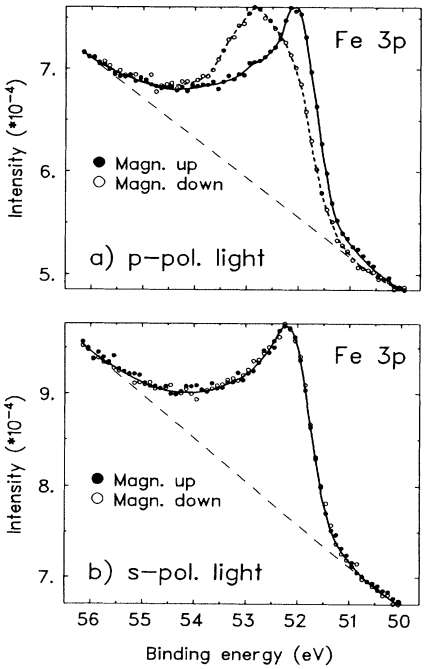

FIG. 2. Fe $3p$ photoemission spectra taken at 90 eV photon energy. (a) Spin-integrated spectra excited with p-polarized light for M up and M down. (b) Spin-integrated spectra excited with s-polarized light under the same geometry as in (a), and for the same magnetization directions.

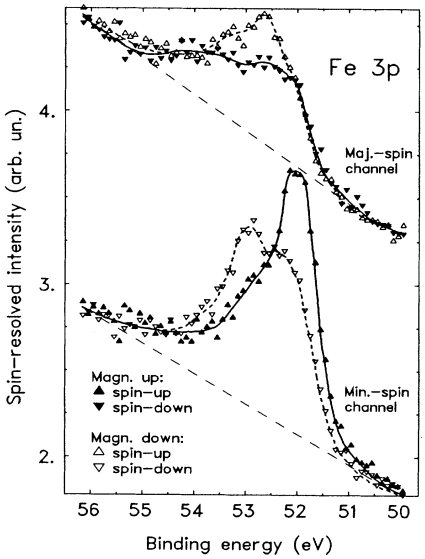

FIG. 3. Spin-resolved energy distribution curves corresponding to the two spin-integrated EDCs in Fig. 2(a) for magnetization parallel ("up") or antiparallel ("down") to the y axis. Maj. and Min. refer to the majority-spin and minority-spin channels, respectively, whereas up and down spin directions are fixed in space (up is parallel to the y axis shown in Fig. 1, down antiparallel).

treatments are by Cherepkov [12] for the atomic case and by Tamura and Feder [8] for photoemission from solids. In our case $\Theta = 16°$, and consequently, by virtue of SO interaction alone, spin polarization in the y direction is expected to occur. This is actually the case in the Fe $3p$ spectrum as can be seen from Fig. 4. This figure shows spin-resolved EDCs derived from the data of Fig. 3 by averaging the spin-resolved EDCs with the same spin orientation (referring to the spatially fixed y axis) over the two magnetization directions. It is clear that these spectra would be obtained from a demagnetized Fe sample with equal numbers of up and down magnetic domains [13]. The main peaks in the spin-resolved EDCs are separated by 0.7 eV. We attribute this to the $3p_{1/2}/3p_{3/2}$ SO splitting which has not been observed before. By comparison with Cu, and considering the fact that SO effects scale with the fourth power of Z_{eff} (the effective nuclear charge) we estimate that the SO splitting in the Fe $3p$ spectrum should be of the order of 1 eV, in fair agreement with the interpretation of the curves in Fig. 4. In the case of Ar $3p$ ionization ($^2P_{1/2}$ final state), a spin polarization of about 10% is found for the 16° angle between the E vector of p-polarized light and the electron momentum [9]. This is of the same order as observed here for the Fe $3p$ peak (15%).

We now can give a qualitative explanation for the magnetization dependence of the spin-integrated EDCs in Fig. 2(a): In the ferromagnet, the exchange interaction of the core hole spin with the valence electrons causes spin polarization in the photoemission spectrum [4,5]. We consider the interplay between the SO and the exchange interactions as the reason for the dependence of the spectra on the direction of the sample magnetization. Because of exchange interaction, the $3p$ spectrum is composed of spin-polarized components where the spin character can be referred to the magnetization direction, which here is either "up" or "down" in space (i.e., parallel or antiparallel to the y axis; see Fig. 1). As described earlier, the SO-induced spin polarization is collinear to the y axis, with a sign determined by the details of the ionization process [8,9,11,12]. Accordingly, the lines with spin polarization originating from exchange are either enhanced or suppressed, depending on whether exchange and SO-induced spin polarizations are parallel or antiparallel. With s-polarized light, of course, the spectra are also spin polarized because of the exchange interaction. However, for symmetry reasons, spin polarization due to SO interaction cannot occur [8]. Therefore, the spectra cannot depend on the direction of magnetization, as we observe in Fig. 2(b). We note also that with p-polarized light, upon full emission angle integration, spin-orbit-induced spin polarization vanishes [9,12] and accordingly, *in angle integrated* photoemission, the $3p$ line shape should not depend on the magnetization direction (up or down).

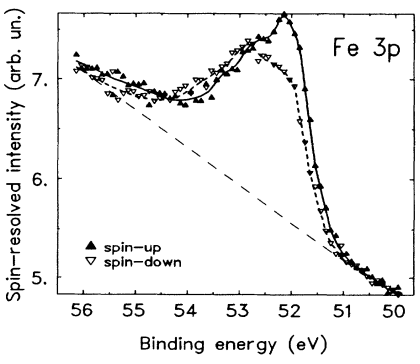

FIG. 4. Spin-resolved Fe $3p$ EDCs determined by spin-orbit interaction for the case of p-polarized light. Spin directions refer to the y axis.

Of course, a theory is required to consider the effects of spin-orbit and exchange interactions *simultaneously* in the core hole photoemission process. The influence of an ordered magnetic moment on the core level spectra excited by linearly polarized light was investigated theoretically by Thole and van der Laan [14]. Their analysis is mainly for localized systems, where the atomic character of the outer d levels which are responsible for the magnetism prevails. There are no angular dependences in their final results, so their results do not relate directly to the experiment presented here. The magnetic linear dichroism predicted by those authors differs qualitatively from the effect demonstrated here, since it manifests itself in a dependence of the photoelectron EDCs on the angle φ between light polarization and magnetization vector ($\varphi = 0°$ and $\varphi = 90°$). That is another effect besides the magnetization *sign* dependence for *fixed* angle ($\varphi = 90°$) that we report on here. However, we also see a dependence of angle-resolved spectra on the relative orientation of electrical vector and magnetization direction which resembles the linear magnetic dichroism effect predicted by those authors. This will be addressed in a forthcoming publication [15].

The linear magnetic dichroism presented here could have practical application in element-specific imaging of magnetic structures. The maximum intensity asymmetry which we observe amounts to 5% at 51.9 eV binding energy. An asymmetry as large as 20% is expected at higher photon energies (250 eV) where the inelastic background is much smaller than in the present experiment. Because of the strong dependence of the $3p$ photoemission spectrum on magnetization for the geometry chosen here, it might even be that the reflectivity for p-polarized light of this energy depends on the sample magnetization direction.

In summary, we have shown for the first time a very strong influence of magnetization direction on spin-integrated photoelectron energy distribution curves obtained with linearly polarized light. By spin-resolved measurements we show explicitly that this is due to the coexistance of spin-orbit and exchange-induced spin po-

larization in the Fe $3p$ spectra. Since the commonly used in-plane synchrotron radiation is linearly polarized, the *linear* magnetic dichroism effect reported here can be exploited most easily for magnetic diagnostic purposes. Furthermore, also the p-polarized component of unpolarized light should cause magnetic linear dichroism in angular resolved photoemission.

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