Circular magnetic dichroism in spin-resolved Fe 3p photoemission

F. U. Hillebrecht, Ch. Roth, and H. B. Rose
Institut für Angewandte Physik, Heinrich-Heine-Universität Düsseldorf, 40225 Düsseldorf, Germany

M. Finazzi and L. Braicovich
Istituto di Fisica, Politecnico di Milano, Piazza Leonardo da Vinci, Milano, Italy
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A photon- and electron-spin-resolved core level photoemission experiment on ferromagnetic Fe is reported. The Fe 3p spectrum shows a circular magnetic dichroism of 40%. We demonstrate that besides the exchange-induced spin polarization the dichroism is accompanied by spin-orbit-induced spin polarization, which depends on the photon helicity but not on sample magnetization. In the spin-integrated as well as spin-resolved data, we observe a strong similarity between circular and linear magnetic dichroism reported by us earlier. This shows a close relationship between the two types of magnetic dichroism.

Magnetic circular dichroism (MCD) in core-level spectroscopy, i.e., the dependence of a core-level spectrum on the relative orientation of magnetization and light helicity, is useful to obtain information on magnetic moments of solids in a chemically specific way.[1,2] In absorption of circularly polarized x rays, the core electron is excited to an unoccupied state immediately above the Fermi level, and there are stringent sum rules, allowing us to extract quantitative information from such spectra.[3] A *photoemission* MCD experiment on a core level by Baumgarten *et al.*[4] showed different spin-integrated Fe 2p spectra for light helicity and sample magnetization either parallel or antiparallel. Thole and Van der Laan[5] (TL) showed for localized magnetic systems that detailed information on electrostatic and spin-orbit interactions as well as on the moments of the magnetization can be obtained from such experiments,[4,6] especially if spin resolution is available. In general, spin resolution allows us to examine the origin of a magnetic dichroism more closely by separating exchange and spin-orbit-induced spin polarizations. Our spin-integrated as well as the spin-resolved results show a close similarity to the recently found effect of magnetic linear dichroism in the angular distribution of photoelectrons (MLDAD).[7,8] Concerning the comparison to the model of TL, it appears that the total spin-orbit-induced spin polarization in the Fe 3p spectrum is finite, in contrast to expectation.

The experiment was performed at the crossed-field-undulator beamline U2 at BESSY, Berlin,[9] using a hemispherical analyzer with a VLEED Fe(100) spin polarimeter.[10] The degree of circular polarization in the third harmonic of the undulator used for 90 eV was $30\pm5\%$;[9,11] the sign of the helicity is not known. The samples were Fe films of 50–100 Å thickness grown on Ag or Au (100) surfaces by electron-beam evaporation from high purity Fe rods. As in our previously reported experiments,[7,8] light is incident under 16° (measured to the surface) nearly parallel to a [100] direction of the Fe surface. The azimuthal angle between light incidence and this direction is 5°. The sample is magnetized along the [100] direction nearly collinear to the light incidence, which is also the quantization axis for the spin polarization measurement. Photoelectrons were collected under normal emission with 0.4 eV energy resolution and 8° acceptance.

Figure 1 shows Fe 3p spectra taken with circularly polarized light for the two sample magnetizations. Both line shapes are different from most of the shapes reported until now[12] for excitation by linearly polarized light, in as far as they show *two* structures, at 52.2 and 52.9 eV binding energy, rather than just a single asymmetric peak. The intensities of the two structures change when the magnetization is reversed: While one of the structures gains intensity, the other one loses. For the opposite helicity we find a virtually identical set of spectra, however with exchanged roles (not shown). As expected,[5] the total intensity is the same in both spectra. The total widths of the 3p spectra, about 1.8 eV, are similar to those seen in previous experiments.[12]

The deviations of shape between these two spectra can be characterized by their difference.[13] The difference spectra for the two helicities (obtained by reversing the sample magnetization) are mirror images of each other. This means that the degree of circular polarization is the same for both helicities within experimental uncertainty, and that CDAD (Ref. 14) effects do not play a role, as expected for coplanar geometry of helicity, magnetization, and electron emission.[15] The difference shows a dispersionlike plus-minus feature between 51 and 55 eV binding energy (BE), which coincides with the apparent base width of the Fe 3p spectrum. The maximum difference is 2.8% of the total intensity at 52.9 eV. For fully circularly polarized light the difference would be 9.3%,[16] and after removal of the secondary electron background a peak difference of $40\pm5\%$ of the total signal is obtained. For an analysis of the absolute strength of the dichroism, e.g., in the framework of the treatment of TL, the specific geometry has to be taken into account: In our angle-resolved experiment, the dichroism is 2.36 times larger than it would be in an angle-integrated measurement,[5d,17]

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while the spectral shape should be unaffected.

To model the different line shapes, one may consider the spectrum in the independent particle picture to be composed of $j=\frac{3}{2}$ and $j=\frac{1}{2}$ one-electron states, which in turn are split up by exchange interaction according to their m_j quantum numbers. As pointed out by Ebert,[18] in first-order approximation the energies of these states increase by a constant amount (the exchange splitting ΔE_{ex}) in the sequence $m_j=-\frac{3}{2},-\frac{1}{2},\frac{1}{2},\frac{3}{2}$ for the $j=\frac{3}{2}$ states, and as $m_j=\frac{1}{2}, -\frac{1}{2}$ for the $j=\frac{1}{2}$ states. In our modeling, the exchange ($\Delta E_{ex}=0.33$ eV) and spin-orbit splitting ($\Delta E_{SO}=0.94$ eV) were adjusted to obtain good correspondence to the experimental spectra. The appropriate spectral intensities for transitions into a structureless εd continuum are governed by Clebsch-Gordon coefficients and the electric dipole matrix elements for circularly polarized light. The dichroism spectrum derived in this way[19] is shown by the full line in Fig. 1(b). It agrees qualitatively with the measured spectrum; however, the negative peak is much stronger than observed in the experiment.

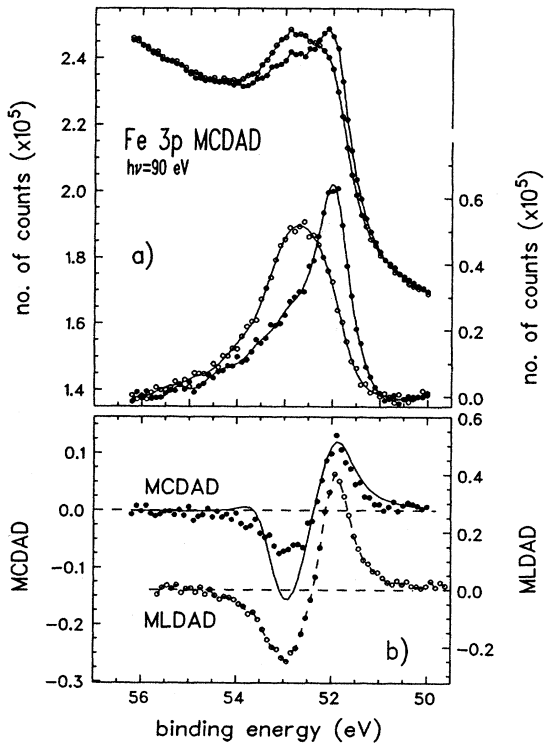

FIG. 1. (a) Spin-integrated Fe $3p$ photoemission spectra taken with circularly polarized light of 90 eV. The spectra correspond to the two possible orientations of helicity and sample magnetization. Equivalent changes are caused by reversing sample magnetization or light helicity. Also shown are spectra corrected for complete circular polarization after removal of secondary background (right scale). (b) Intensity difference for circular (full circles, left scale) and linear magnetic dichroism (MLDAD, dashed line and empty circles, right scale). The full line through the MCDAD data points is a result of a simulation. Vertical scales are numbers of counts divided by 10^5.

For comparison, the lower panel of Fig. 1 shows the magnetic dichroism observed with p-polarized light and switching the magnetization between the two directions normal to the plane of electric field and electron emission (MLDAD).[7,8] The line shapes of the circular and linear dichroism curves are very similar. This strong similarity is a hint for an intimate relationship between these two forms of dichroism. For the linear case the peak dichroism amounts to 4.7% of the average intensity (including background), which is half as strong as the circular dichroism extrapolated to complete circulate polarization.

In Fig. 2 the MCD data with spin resolution are shown. In this figure the spin polarization of the secondary background (about 10% under the conditions of our

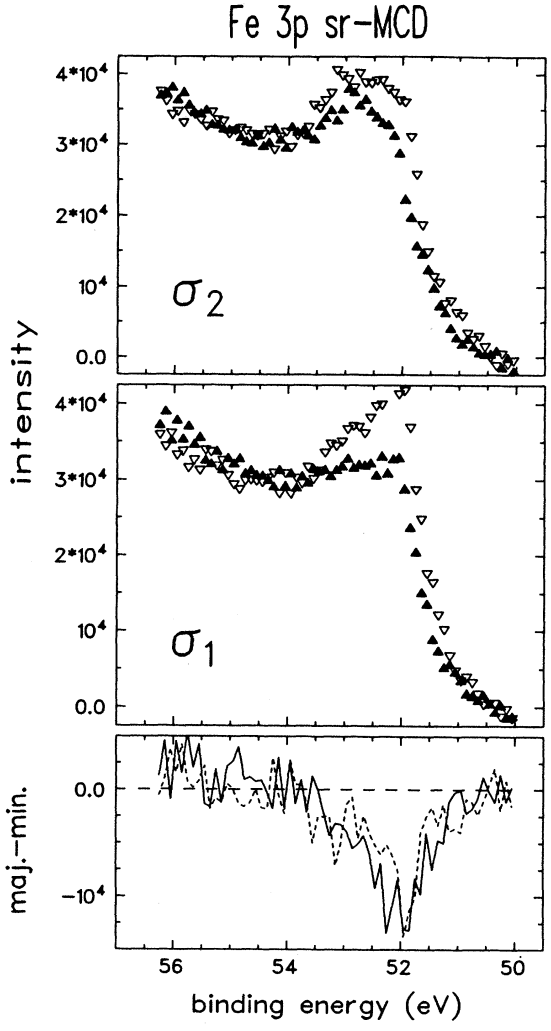

FIG. 2. Spin-resolved Fe $3p$ photoemission spectra excited by circularly polarized light for the two possible relative orientations of light helicity and sample magnetization (filled triangles pointing up: majority, empty triangles pointing down: minority spin). A 10%-polarized background was suppressed. Lowest panel shows difference between majority and minority spin intensities for σ_1 (full line; filled circles in Fig. 1) and σ_2 (dashed; empty circles in Fig. 1). Vertical scale is number of counts.

experiment) was suppressed so that majority and minority spin energy distribution curves have the same base. Reversing the light helicity has a similar effect on the spin-resolved spectra as it has on the spin-integrated spectrum. The general features are relatively sharp $3p$ peaks in both spin channels for σ_1 helicity. When the helicity is reversed, these sharp structures get rounded, and intensity is transferred to higher binding energy. This applies to both spin channels. In the lowest panel of Fig. 2 the absolute intensity difference of majority and minority spin intensities is shown.[13] These differences are negative, reflecting the fact that the $3p$ peaks are dominated by minority spin character. We note that the spin polarization is larger for σ_1 than for σ_2 helicity.

The top panel of Fig. 3 shows the spin-resolved spectra combined such that the sample magnetization is averaged out:

$$I_{\text{up}} = 1_{\sigma 1,\text{maj}} + I_{\sigma 2,\text{min}} \ , \quad I_{\text{down}} = I_{\sigma 1,\text{min}} + I_{\sigma 2,\text{maj}} \ .$$

These spin-resolved data represent the spectra for spin either parallel or antiparallel to the photon helicity. As these spectra are different, it is clear that there is a spin polarization which is not related to magnetism, i.e., exchange interaction. The cause of this spin polarization is the spin-orbit interaction. From a number of investigations[20–22] of photoionization, e.g., of rare gases or alkali vapors, the properties of this type of spin polarization are established. For any given geometry and negligible spin-orbit interaction in the continuum final state, the spin-orbit-related polarization should vanish if the spin-orbit splitting is not resolved. To check for this behavior, we show in Fig. 3(b) the difference of these two spectra, which is discussed as spin-orbit (so) spectrum by TL (denoted as $I_{11}{}^{5a,b}$). This spectrum represents the spin character of the $3p$ spectrum with regard to photon and electron spin being either parallel or antiparallel to each other. According to TL, positive and negative peaks in the spin-orbit spectrum correspond to core hole states with high or low total angular momentum j, and for excitation out of a closed shell the integrated intensity should be zero. In our experiment, its integral does not vanish: In the region from 50 to 54 eV BE the so spectrum shows a broad negative structure.[23]

We also show the result of our model calculation for the spin-orbit spectrum.[19] The individual states are shown by bars in Fig. 3(a). The overall spectra obtained by taking lifetime broadening and instrumental resolution into account show some resemblance in shape with the experimental spectra. However, experimentally the two spectra have different integrated intensities, while the calculated ones have equal intensities. A finite overall spin-orbit polarization as evidenced by these data can be caused by spin-orbit interaction in the photoelectron continuum, which can have a significant effect particularly when the total cross section has a minimum, as in the Fano effect.[20–22] This contribution has been neglected in our model, as well as in the treatment of TL.

There is qualitative agreement between the present result and our result for the linear magnetic dichroism observed with linearly p-polarized light in chiral geometry [MLDAD (Ref. 8)]. The spectrum in Fig. 3(c) shows the intensity difference incurred by spin-orbit-induced spin polarization for this geometry (the difference of the two spectra in Fig. 4 of Ref. 8). In qualitative agreement with the MCD result, the so-induced spin polarization is dominated by one feature between 51 and 53 eV BE. In both cases, for circular as well as for linear dichroism, spin-orbit interaction leads to a spin polarization *independent* of the spin polarization caused by exchange interaction with the spin-polarized valence electrons of the ferromagnet. In the case of the linear dichroism this polarization is normal on the reaction plane, and the dichroism is observed when the magnetization is in this direction.[8] For

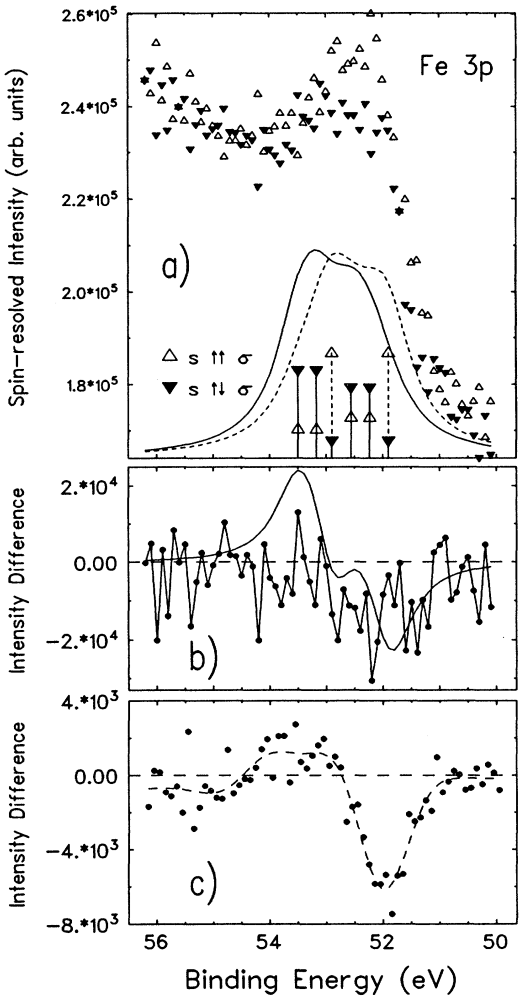

FIG. 3. (a) Spin-resolved Fe $3p$ photoemission spectra ($h\nu = 90$ eV) corresponding to spin parallel and antiparallel to light helicity (extrapolated to complete circular polarization). Bars show result of a simulation, which leads to the spectra shown by dashed and full lines. Exchange splitting is 0.33 eV, spin-orbit-splitting 0.94 eV, lifetime broadening 1.0-eV full width at half maximum. (b) Difference of the two spin-resolved spectra, i.e., the spin-orbit (so) spectrum according to TL; line shows result of simulation. (c) Analog of so spectrum derived from MLDAD. Dashed line is a spline fit through the data. Vertical scales are numbers of counts. Scale in (c) is not related to other scales.

circularly polarized light we observe a dichroism when the magnetization is (nearly) collinear to the light propagation vector. Here, the spin polarization imposed by spin-orbit interaction has a component parallel to this direction. Depending on emission angle, there may also be other components of the so-induced spin polarization,[21] connected with the appearance of magnetic dichroism in other geometries.[17,15,24] Both for circular and linear dichroism the matrix elements for transitions to $l+1$ and $l-1$ final states play a role, where l is the orbital momentum of the core level. For MLDAD, also the phases of final-state waves with orbital character $l+1$ and $l-1$ are of importance.[24] Consequently, the energy and angle dependences of the two types of dichroism will be different.

We have reported a photon-resolved and electron-spin-resolved core-level photoemission experiment on the $3p$ level of ferromagnetic Fe. The maximum measured circular dichroism of 2.8% of the total intensity extrapolates to 40% for complete circular polarization and background removed. Spin analysis has allowed us to measure the spin-orbit spectrum, representing the spin-orbit-induced spin polarization. Contrary to expectation, integration of this spectrum over the energy range of the Fe $3p$ peak does not yield zero. Within the present understanding, these results suggest that the final states with different total angular momentum j are not contributing according to their statistical weight to the Fe $3p$ photoemission peak. A remarkable finding is the apparent equivalence of circular and linear dichroism, which is clearly evident in the spin-integrated data, and (due to technical limits) to a lesser degree in the spin-resolved results. Since the report of MLDAD,[8] a number of theoretical treatments have shown that the two kinds of dichroism are in fact equivalent,[15,24,20] and the results reported here constitute the most direct experimental proof of this.

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