



MULTIPLY STRUCTURE IN HIGH-RESOLUTION SPIN-POLARIZED Fe 2p PHOTOEMISSION EXCITED BY CIRCULARLY POLARIZED RADIATION

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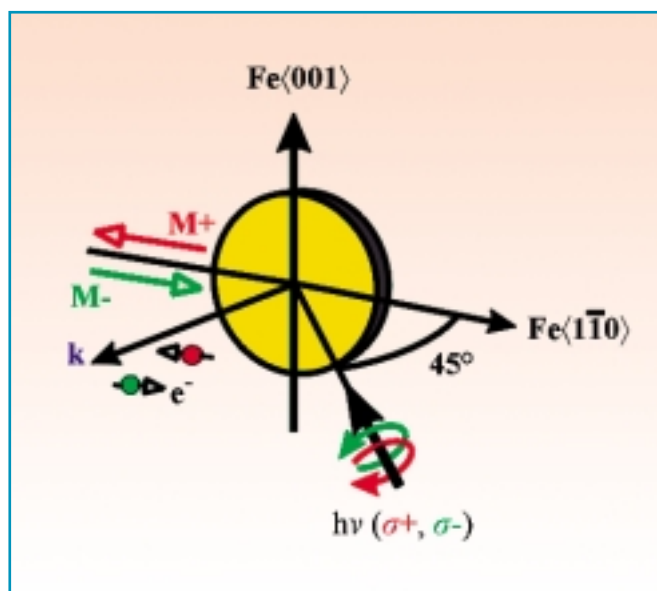
Magnetic circular dichroism in 2p excitations of Fe was studied by high-resolution spin-resolved core-level photoemission. The highly differential experiment reveals richly-structured spectra which allow the first direct identification of four constituent multiplets in the $j = 3/2$ final state.

Core-level photoemission spectra of magnetic materials may carry information on the magnetic ground state in two ways: firstly, via the spin polarization of the photoelectrons, and, secondly, by the occurrence of magnetic dichroism. Experimental and theoretical studies addressing these issues for photoemission from the 3d ferromagnets and rare earths have been reported by a number of groups. Electron spin analysis commonly reduces the photoelectron count rate by about three orders of magnitude, which limits spin-resolved experiments to high-performance beamlines at third generation synchrotron sources, such as the ESRF. It is clear that a spin and angle-resolved experiment is the most differential photoemission experiment possible, and therefore may reveal more information on the spectral lineshapes than can be obtained from other experiments. While the photoemission spectra of localized systems, e.g. the rare earths, can be analyzed in a straightforward manner using an atomic approach, systems in which the magnetic moment is carried by delocalized d electrons are far more complicated. For such systems it is not clear to what extent atomic effects prevail, or whether the spectra can be explained within a one electron model. This issue is best addressed in the context of the 2p photoemission spectra of the 3d ferromagnets, because - in contrast to the 3p level which is easily accessible experimentally - the spin orbit splitting is large compared to other interactions, which should simplify the analysis.

For the example of the 2p photoemission lines of the 3d ferromagnets, it is clear from the relatively large line widths observed in conventional photoemission spectroscopy that multiplet interaction must be present in the 2p spectra of 3d ferromagnets [1, 2]: the linewidths are significantly larger than in comparable materials to be attributable only to lifetime broadening. However, to separate the multiplet components experimentally is difficult if not impossible since the splitting between them is comparable to the lifetime broadening. Our goal was to elucidate the detailed spectral features of the 2p photoemission spectrum in a highly differential experiment, combining magnetic dichroism and spin analysis.

As a general rule, magnetic dichroism occurs in photoemission if there is a finite spin polarization caused by spin-orbit interaction along the direction of magnetization. This makes a spin polarized study of magnetic dichroism particularly desirable because, from such a study, one can separate exchange and spin-orbit effects in the photoemission spectra. This was the motivation for our high-resolution study of the Fe 2p level by spin resolved photoemission excited by circularly polarized light. Our results show structures which unambiguously demonstrate the presence of discrete final states in the photoemission spectrum, and allow, for the first time, a direct determination of their energy positions and relative intensities. This

Fig. 1:
Geometry of the experiment and coordinate system with electron wave vector k , magnetization M , and measured component of spin-polarization.

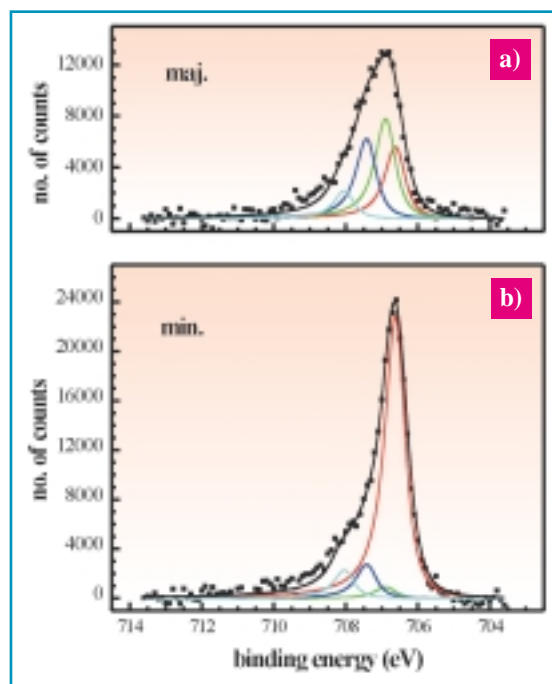


information is vital for an adequate description of core-level photoemission spectra of 3d magnetic systems.

The experiments were carried out at ESRF beamline ID12b, using our photoemission spectrometer equipped with an Fe VLEED spin polarimeter [3]. This method of spin analysis utilizes the spin-dependent reflectivity of a magnetized Fe(100) surface for low energy electrons, which offers a roughly 10 times higher figure of merit than other methods of spin analysis. The samples were grown as ultrathin films (about 20 ML) of Fe on W(110). The overall energy resolution was 0.45 eV. The geometry of the experiment is shown in Figure 1: circularly polarized light from beamline ID12b impinges at 45° onto the sample, and the photoelectrons emitted from the sample are collected in normal emission. The sample is magnetized in the surface plane, in the direction at which the incident light is projected onto the surface. This is also the direction along which the spin polarization is analyzed.

Figure 2 shows spin-resolved Fe $2p_{3/2}$ photoemission spectra excited by 850 eV circularly polarized radiation for fixed light helicity and two opposite sample magnetizations. The background of secondary electrons was subtracted in the usual manner from the spectra. The magnetic dichroism (not shown), i.e. the difference between the two spectra normalized to their sum, amounts to 20%. Since the sample magnetization and the light helicity are at 45°, the observed dichroism extrapolates to an intrinsic dichroism of 28% for this photon energy. In the spin-resolved data, all lineshapes are different, and the individual spectra show characteristic features. To analyze these lineshapes, the spectra were fit with as many Doniach Sunjic lines as required to describe the data. We assume that the spectra are composed of lines with identical binding energies (BE's), with only the intensities changing between the spin channels or with magnetization reversal. The characteristic features in the spectra allow the determination of the number of discrete states present. It is evident from the spectra, that the minimum number of lines to describe the spin-resolved spectra is four: The majority spectrum, Figure 2a, clearly consists of three lines. However, the minority spectrum, Figure 2b, shows a peak at a binding energy lower

Fig. 2: Spin-resolved Fe $2p_{3/2}$ spectrum for magnetization up. Dots show experimental spectrum after background subtraction, lines show fits to the spectra by four Doniach-Sunjic lines. The constituent spectra are shown without applying the experimental broadening of 0.45 eV.



than the lowest binding energy peak evident in the majority spectrum. This can only be described by a fourth line at the appropriate energy. The fits were obtained by an iterative procedure, varying the binding energies of the constituent lines in all spectra (the two spectra shown plus two more for the opposite magnetization) in the same way. The Lorentzian broadening of 0.37 eV, Gaussian broadening of 0.45 eV, representing the experimental resolution, and the asymmetry index $\alpha = 0.2$ were chosen to give an optimum fit to all spectra. The binding energies, relative intensities, and spin polarizations are given in Table 1. The data show that the energy positions of the sublevels are not equidistant. While the splitting between the three higher binding energy lines are 0.62 and 0.53 eV, respectively, the splitting between the two lowest energy lines is only 0.27 eV. This splitting can also be read directly from the spectra in

Figures 2a and 2b.

Overall the fits describe the observed spectra very well. In contrast to previous modeling studies, this analysis is based exclusively on the experimental evidence. Some discrepancies are still present in the region above 708.5 eV binding energy, where the experimental spectra for magnetization down show a tail which is not described by the modeling. The magnetic dichroism curves - also for the linear dichroism - consistently show a broad peak in this energy region. This satellite probably originates from the low spin final states which in the atomic limit are spread out over at least 20 eV. Nevertheless, one finds in the magnetization up spectrum that the leading line is very strongly polarized, about -63% (- for minority, derived from the intensities in table 1). Also the second line for magnetization up shows a large polarization, but now of opposite sign, +77%. For the polarization integrated over the

Table 1: Binding energies and intensities of the components of spin-resolved Fe $2p_{3/2}$ spectra excited by circularly polarized light. The last line gives the overall spin polarization (- means minority spin type polarization).

| BE (eV) | MAG + | | |
|-------------------|-------------|-------------|--------------|
| | maj | min | pol (%) |
| 706.58 | 10.5 | 46.9 | -63.3 |
| 706.85 | 14.6 | 1.9 | +77.2 |
| 707.38 | 11.7 | 5.6 | +35.1 |
| 708.00 | 4.1 | 4.7 | -6.6 |
| sum | 40.9 | 59.1 | -18.1 |
| | 100 | | |
| exp. total | 43.3 | 59.9 | -16.1 |



spectrum, the polarizations cancel partially, so that the overall polarization is -16.1%. So far, all core-level spectra measured show minority spin polarization within the peak region, consistent with the present finding.

Next we turn to the spin polarizations related to spin-orbit and exchange interaction. Both effects generate a photoelectron spin polarization by themselves, i.e. without the other one present, and they may enhance or suppress the spin polarization observed in a spectrum excited by circularly polarized light. This is evident in the different spin polarization (integrated over the photoemission peak) found with reversed sample magnetization or light helicity. The exchange polarization is obtained by classifying the spin-resolved spectra with respect to majority and minority. The remaining polarization of -13% does not contain the spin-orbit effect, and vanishes when the temperature is raised above the Curie point, i.e. it is related to the magnetism of the sample. It is caused by the exchange interaction of the core hole spin and the magnetically polarized valence electrons. In general, photoelectron spectra are influenced by diffraction of the photoelectrons by the crystalline surroundings of the emitting atom. However, we have shown in other spin-resolved dichroism studies that the exchange polarization is affected only very weakly by diffraction.

The spin-orbit polarization is obtained by classifying the spin-resolved spectra with respect to the spin direction imposed by the light helicity, i.e. by averaging out the effect of magnetization reversal. Since the polarizations observed for the two magnetizations differ, this would yield a finite result even if the sample was demagnetized. Therefore, it is an effect

caused by temperature-independent spin orbit (so) interaction, which can also be observed on non-magnetic materials, e.g. Cu [4]. In our experiment we found an so-polarization of -3%. Since magnetic dichroism and spin orbit polarization are closely related [5], there should be a sizeable so polarization. As a guide to the order of magnitude one may use the so polarization found in Cu metal as the basis for an estimate. This suggests an expected so-polarization of 15 to 20% for Fe under our experimental conditions. Photoelectron diffraction (PED) is unlikely to be the cause for the apparent disparity between magnetic dichroism and spin-orbit polarization since we know from linear dichroism [6] that PED scales spin-orbit polarization and magnetic dichroism in the same way depending on emission direction. Analysis of the data taking into account photoelectron diffraction in a multi-scattering approach is currently under way.

The results presented here show that the large width of the Fe 2p photoemission spectrum arises from the presence of discrete final states, which hitherto have not been observed unambiguously. The fact that *four* such final states can be identified is in agreement with a one electron model [7] in which the magnetically oriented valence electrons provide an effective spin field which splits the $j = 3/2$ states according to their m quantum number. However, the energy splittings and intensities cannot be described satisfactorily by such a model. Also, satellite states which are evident in the tails on the high binding energy sides of the spectra are (at present) beyond such a model. Interestingly, the lifetime broadenings of the majority and minority states appear to be similar. A

non-uniform splitting can be explained if the spin field is comparable to the spin orbit interaction (see figure 5 in ref. [5]). While this is plausible for the 3p level, the spin orbit interaction for the 2p shell is far too large for this explanation to hold [5, 8]. It is hoped that the present data will stimulate further theoretical investigations, which will account for sublevel energies and intensities. ■

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