# Magnetic dichroism in photoemission as a spin-resolving probe for electronic correlations

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Magnetic dichroism with unpolarized x rays in angle-resolved photoemission has been systematically investigated for the $2p$ core levels of Fe, Co, and Ni. In contrast to Fe $2p$, we find the dichroism in the Ni $2p$ spectra to be mainly governed by electronic correlations. The fine structure observed in the Co $2p$ spectrum indicates a distinct influence of many-body interactions also in this material. [S0163-1829(96)50446-7]

Electronic interactions govern many phenomena in solid-state physics. Ferromagnetism, for instance, is a direct consequence of spin-dependent electronic correlations. Besides their influence in the ground state, correlations are very important for understanding electron spectroscopy results. These techniques always probe the many-electron system, even if the results may often be interpreted in the single-particle limit. A famous example for the inadequacy of the single-particle picture is photoemission from Ni. Although a density functional approach can reproduce the average exchange splitting found in the experiment by a proper choice of the exchange correlation potential,[1] it fails to explain the photoemission satellite structures found,[2] yet another manifestation of electronic correlations. Their interpretation requires suitable many-electron formalisms, for example, a Hubbard-type approach as has been successfully applied to Ni.[3] A narrow $d$ band in Ni—as compared to Fe and Co—is held responsible for the formation of correlation-induced spectral features. This bandwidth argument leads to an exciting aspect if surfaces and thin films are considered. It is well-known that the smaller number of nearest-neighbor atoms at the surface causes a dehybridization of the wave functions and thereby a band narrowing. A similar effect happens in monolayer films as a consequence of the reduced dimensionality, as long as hybridization with the substrate is negligible. As pointed out recently by Chen,[4,5] many-particle effects may thus be enhanced in systems of reduced dimensions.

Magnetodichroic phenomena in angle-resolved photoemission appear for various experimental situations.[6–9] Previous studies have centered on the role of experimental geometry, light polarization, and excitation energy. The question of how the presence of electronic correlations will affect the magnetodichroic spectra remained unanswered. In this paper we demonstrate the strong influence of spin-dependent electronic correlations on the magnetic linear dichroism in $2p$ photoemission. In order to make contact to explicitly spin-resolved results, the dichroism was measured with unpolarized x rays. On the one hand, the Fe $2p$ spectra are similar to those observed earlier with circularly polarized light[6] and can be understood within a single-particle picture. On the other hand, we find the highest magnetodichroic signal for Ni $2p$ in the 6 eV satellite. In this case the magnetic dichroism is clearly dominated by correlation effects. The fine structure in the Co $2p$ spectra from a 5 monolayer (ML) thick film shows evidence for a many-body satellite located $\sim$4 eV below the main $2p_{3/2}$ emission line.

Of particular interest when exciting with unpolarized light is the so-called linear magnetic dichroism in the angular distribution of photoelectrons (LMDAD). The effect is observed if the magnetization **M** is oriented perpendicular to the reaction plane spanned by the directions of light incidence (**q**) and electron emission (**k**) (Fig. 1). LMDAD results from an interference of competing excitation channels, involving matrix elements with the components of the electric field **E** parallel to the reaction plane and parallel ($E_x$) and normal to the surface ($E_z$), respectively.[10–12] This view was confirmed by an LMDAD study of the Co $3p$ levels.[13] In the reference frame of Fig. 1, unpolarized light corresponds to an incoherent mixture of ($E_x$,$E_z$) and $E_y$ components. Because $E_y$ does not give rise to a dichroism upon reversing **M** (but contributes to the intensity), LMDAD shows up even with *unpolarized* light.[8,9] The dichroic signal, however, will be a

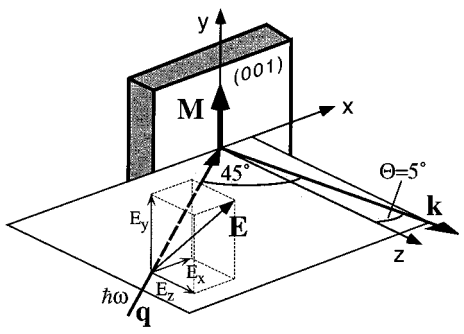


FIG. 1. Geometry of the photoemission experiment indicating the orientations of sample magnetization **M**, light incidence **q**, electron wave vector **k**, and electric field components ($E_x$,$E_y$,$E_z$).

 

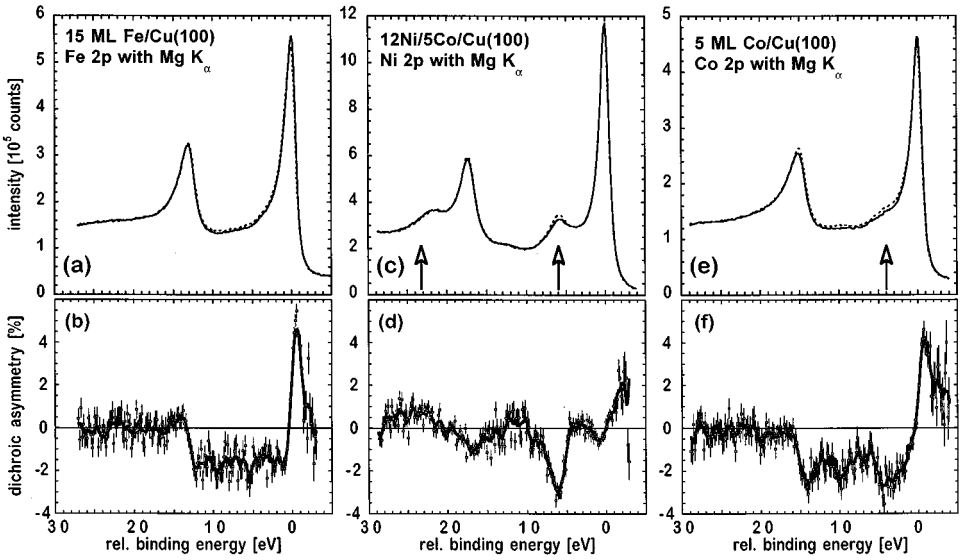


FIG. 2. Photoelectron intensity distribution curves (a) and magnetic linear dichroism signal (b) for the excitation of the Fe $2p$ core levels with unpolarized Mg $K_\alpha$ radiation. Panels (c) and (d) give the corresponding results for Ni $2p$, and panels (e) and (f) those for Co $2p$ (arrows mark the position of correlation-induced satellites).

factor of 2 smaller than for linearly polarized light with $\mathbf{E}\|(x,z)$.

Our experiments were carried out in a UHV two-chamber system (base pressure $1\times10^{-8}$ Pa). The preparation chamber houses facilities for low-energy electron diffraction (LEED), Auger electron spectroscopy (AES), and thin film growth. The epitaxial growth of the films was monitored by medium energy electron diffraction.[14] Fe, Co, or Ni films were deposited by means of electron beam evaporation sources onto a Cu(100) template held at room temperature. After characterizing the films with respect to crystalline structure and cleanliness, the sample was transferred into the adjacent analysis chamber containing a hemispherical spectrometer and a Mg $K_\alpha$ x-ray source (overall energy resolution of the system $\Delta E\leqslant1.5$ eV). The spectra were taken as a sequence of individual runs in a slight off-normal emission geometry [$\Theta=5°$ (Ref. 15)], with the angle between $\mathbf{q}$ and $\mathbf{k}$ being 45°. Prior to each run the sample was remanently magnetized by a current pulse through a magnetization coil. Before, during, and after the photoemission experiments, the magnetic state of the sample was checked by means of the magneto-optical Kerr effect.

The x-ray source delivered mainly Mg $K_{\alpha_{1,2}}$ radiation ($\hbar\omega=1253.2$ eV), with a small fraction of higher-energy satellites. The photoemission contributions from the most prominent ones, $K_{\alpha_3}$ and $K_{\alpha_4}$, have been removed by subtracting appropriately weighted Doniach-Šunjić lines from the measured spectra.

The Fe $2p$ spectrum [Figs. 2(a) and 2(b)] was obtained from a 15-ML-thick film. This coverage ensures an in-plane remanent magnetization of Fe/Cu(100).[16] The spectra recorded for opposite magnetization directions [solid and broken lines in Fig. 2(a)] reveal a small but distinct difference in both peak intensity and energy position (binding energies $E_b$ refer to a relative scale, with its origin set to the average maximum of the $2p_{3/2}$ peak of both spectra). The apparent shift between the $M=1$ and $M=-1$ spectrum is of opposite sign for the $2p_{3/2}$ and $2p_{1/2}$ lines. This gives rise to the characteristic dichroic signal in Fig. 2(b), with a plus/minus feature at $2p_{3/2}$ and a minus/plus feature at $2p_{1/2}$ [we refer to the dichroic signal as an intensity asymmetry $A(E)$, i.e., the difference of the intensity spectra taken at opposite magnetization directions divided by their sum]. The shape of the asymmetry spectrum resembles closely those observed with circularly[6] and linearly polarized light.[17]

*Circular* magnetic dichroism in Fe $2p$ photoemission has been successfully interpreted within a single-particle model, assuming the core levels in the ground state to be split according to their azimuthal quantum number $m_J$.[18] This analysis traced the positive (negative) sign of the $2p_{3/2}$-related asymmetry $A_{3/2}$ back to emission from predominantly minority (majority) spin states, leading to down (up) spin photoelectrons (for the magnetic dichroism in the $2p_{1/2}$ peak the situation is just reversed). In a short form, we can write this relationship as

$$\mathrm{sgn}\{A_{3/2}(m_J)\}=-\mathrm{sgn}\{\langle\sigma_{3/2}(m_J)\rangle\}, \tag{1}$$

with the spin expectation value $\langle\sigma_{3/2}(m_J)\rangle$ of the sublevel $m_J$. A qualitative interpretation of the data in Fig. 2(b) may expand on these results. The matrix elements for transitions from the individual $m_J$ levels will be different in the case of unpolarized light, of course, but the sequence of levels will be the same as in Ref. 18. The relationship between the sign of the dichroic signal and the photoelectron spin character should therefore still follow Eq. (1). Further independent support for this assignment comes from recent spin-resolved experiments with unpolarized[19] and linearly polarized light.[17]

The Ni $2p$ photoemission results were obtained from a 12-ML Ni film grown on a 5-ML Co buffer layer.[20] Each main photoemission peak in the intensity spectra is accompanied by a satellite located at ~6 eV higher binding energy [Fig. 2(c)]. This well-known correlation-induced feature is attributed to the screening of the $2p$ core hole.[21] The effect of electronic correlations on the magnetic dichroism, however, is even more dramatic [Fig. 1(d)]. The asymmetry in the $2p_{3/2}$ and $2p_{1/2}$ lines is drastically reduced to a value $<1\%$. The largest asymmetry is found in the satellites, being negative (positive) for the $2p_{3/2}$ ($2p_{1/2}$) associated features. Using Eq. (1) one finds that the satellite corresponds to ma-

jority spin photoelectrons. This finding confirms the results from explicitly spin-resolving experiments on the Ni core levels.[21]

How can we understand the asymmetry spectrum, in particular, the strongly reduced magnetic dichroism in the main emission lines? Starting from the single particle point of view, we recall that LMDAD involves a phase shift difference $\delta=\delta_0-\delta_2$ between the two interfering final state channels with $\ell=0$ and $\ell=2$, whereby the dichroism scales with $\sin\delta$.[22] Calculating the radial matrix elements and phase shifts in Fe, Co, and Ni for $p\rightarrow s$ and $p\rightarrow d$ transitions, we obtained nonzero values for both $\delta$ and [using Eq. (19) in Ref. 22] for the LMDAD signal. Thus our experimental finding is *not* related to the trivial case $\delta=0$. Still within the single-particle picture we may also consider the scattering of the outgoing photoelectron. Experiments have shown that crystalline effects may cause a pronounced angular variation of the magnetodichroic signal and can even change the sign of $A(E)$.[23] With the electrons having comparable kinetic energy, however, main photoemission lines *and* satellites should be affected in a similar way, in contrast to our experimental observation. The single-particle picture thus clearly fails to explain the reduced dichroism in the main lines, not to mention the satellites.

We now turn to the many-particle point of view. A description of many-body effects in core-level photoemission often takes recourse to atomic multiplet treatments. Configuration interaction theory, for instance, predicts the ground state of ferromagnetic Ni—in terms of the $3d$-electron occupancy—to be a superposition of $d^8$, $d^9$, and $d^{10}$ atomic configurations.[24] Among those $d^{10}$ corresponds to a filled $d$ shell with no net magnetic moment. The core-hole screening causes a redistribution of the charge density in the vicinity of the core hole.[21] In addition, the electrostatic interaction between core hole and electrons renormalizes the binding energies. Thus, the $d$ electrons become more strongly bound and a small fraction of the previously unoccupied density of states shifts below the Fermi level $E_F$. As the Ni majority spin states are practically fully occupied, this shift reduces the number of unoccupied minority spin states and thereby directly the magnetic moment. In the above picture, the core-hole screening changes the weight of the atomic configurations, with basically only $d^9$ and $d^{10}$ left in the final state.[21] The main photoemission line is then associated with $3d^{10}$, whereas the $3d^9$ contribution is responsible for the satellite. This atomic model qualitatively explains the experimental findings, but completely neglects the itinerant character of the $3d$ electrons in Ni, being important for extraatomic contributions to the screening. Itinerancy is taken into account in a so-called small cluster model, whereby a good agreement with spin-resolved Ni $2p$ photoemission data was found.[25] A magnetic *circular* dichroism spectrum calculated in the same paper[25] agrees surprisingly well with our data obtained with unpolarized light: the largest dichroism is found in the satellite peaks, whereas it vanishes for the main lines. The latter prediction is at variance with our observations of a residual dichroism. The difference may be due to the excitation with unpolarized instead of circularly polarized light or the different geometry assumed in the theory in Ref. 25.

The situation for the Co $2p$ photoemission by contrast differs from both Fe and Ni. The overall shape of the intensity spectra from a 5-ML fcc-Co film [Fig. 1(e)] resembles that of Fe $2p$, but with the distinct difference of an additional weak shoulder at $E_b\sim 4$ eV. The feature becomes clearer when looking at the asymmetry spectrum [Fig. 1(f)]. It is related to a marked negative excursion in $A(E)$ peaking at $E_b\simeq 4.5$ eV. The seemingly larger width of the $2p_{1/2}$ peak—as compared to iron—also suggests the presence of some (unresolved) additional feature in the high binding energy flank. In analogy to the discussion of the Fe and Ni results, we are therefore led to interpret these extra features as correlation-induced satellites of predominantly majority spin character.

The issue of many-body satellites in the Co $2p$ spectra is still controversely discussed. In their recent spin-resolved work, Klebanoff *et al.* observed no indication for such satellites.[26] This contradicts earlier results from conventional XPS by Raaen,[27] who reported an extra spectral feature located at $E_b\approx 4.5$ eV, in agreement with our own finding. Observations made from different systems, however, may not be directly comparable. The energetic position and spectral weight of core-level many-body satellites may depend sensitively on details of the valence electronic structure, which in turn may change with the crystalline structure and morphology of a sample. In Ref. 26 polycrystalline Co films thicker than 60 Å were grown on a Co-based met glass, whereas Ref. 27 states neither film thickness nor substrate. Our Co films on Cu(001) have a well-defined fcc-like structure with a slightly tetragonally compressed vertical interlayer spacing.[28] A thickness of 5 ML in our experiments was chosen, because both the magnetic properties and the electronic structure of this system are well known.[14,29]

Strong support for the existence of correlation-induced spectral features in Co comes from theory. For a Co monolayer on Cu(001) theoretical investigations find majority spin satellites (located at about 4.5 and 9 eV below $E_F$) in valence band photoemission spectra.[5] We note that the first value comes close to our results and those in Ref. 27. The formation of these satellites is supported by a strong $d$-$d$ correlation and a narrowing and dehybridization of the Co $d$ bands. In fact, as soon as a Co$3d$/Cu$4s$ hybridization at the interface is introduced, the spectral weight of the satellites is substantially reduced.[5] In the absence of calculations for thicker films the above prediction raises two interesting questions. The first concerns the influence of a surface where also band narrowing and electronic dehybridization takes place. For thicker films the influence of the $s$-$d$ interfacial hybridization onto the surface electronic structure can be neglected. One may speculate that under these circumstances the strong $d$-$d$ correlation may still lead to spectral satellites strong enough to be observed experimentally. The second question addresses the role of the film's structure. A tetragonal distortion of an fcc lattice, for instance, reduces the symmetry and will change details of electronic states, in particular, close to $E_F$.[29] In order to clarify the influence of these effects on electronic correlations and the formation of many-body satellites, further theoretical and experimental efforts are needed.

In summary, we have shown that magnetic linear dichroism in the angular distribution of photoelectrons can be used as an alternative technique in order to investigate spin-dependent correlation effects in core-level photoemission.

The Ni $2p$ spectra, in contrast to Fe $2p$, are strongly affected by electron-electron interactions, which dominate the magnetic dichroism and cannot be understood in a single-particle picture. In the Co $2p$ results we found additional spectral features in both the intensity distributions and magnetic dichroism spectra, which are tentatively attributed to the presence of correlation-induced satellite peaks.

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[1] H. Eckardt and L. Fritsche, J. Phys. F **17**, 925 (1987).
[2] F.J. Himpsel, J.A. Knapp, and D.E. Eastman, Phys. Rev. B **19**, 2919 (1979).
[3] W. Nolting, W. Borgiel, V. Dose, and T. Fauster, Phys. Rev. B **40**, 5015 (1989).
[4] Changfeng Chen, Phys. Rev. Lett. **64**, 2176 (1990).
[5] Changfeng Chen, Phys. Rev. B **48**, 1318 (1993).
[6] L. Baumgarten, C.M. Schneider, H. Petersen, F. Schäfers, and J. Kirschner, Phys. Rev. Lett. **65**, 492 (1990).
[7] Ch. Roth, F.U. Hillebrecht, H. B. Rose, and E. Kisker, Phys. Rev. Lett. **70**, 3479 (1993).
[8] F.U. Hillebrecht and W.-D. Herberg, Z. Phys. B **93**, 299 (1994).
[9] M. Getzlaff, Ch. Ostertag, G.H. Fecher, N.A. Cherepkov, and G. Schönhense, Phys. Rev. Lett. **73**, 3030 (1994).
[10] D. Venus, Phys. Rev. B **49**, 8821 (1994).
[11] B.T. Thole and G. van der Laan, Phys. Rev. B **49**, 9613 (1994).
[12] N.A. Cherepkov, Phys. Rev. B **50**, 13 813 (1994).
[13] W. Kuch, M.-T. Lin, W. Steinhögl, C.M. Schneider, D. Venus, and J. Kirschner, Phys. Rev. B **51**, 609 (1995).
[14] C.M. Schneider, P. Bressler, P. Schuster, J.J. de Miguel, R. Miranda, and J. Kirschner, Phys. Rev. Lett. **64**, 1059 (1990).
[15] M. Salvietti, X.Y. Gao, C.M. Schneider, and J. Kirschner (unpublished).
[16] J. Thomassen, F. May, B. Feldmann, M. Wuttig, and H. Ibach, Phys. Rev. Lett. **69**, 3831 (1992).
[17] F.U. Hillebrecht, Ch. Roth, H.B. Rose, W.G. Park, E. Kisker, and N.A. Cherepkov, Phys. Rev. B **53**, 12 182 (1996).
[18] H. Ebert, L. Baumgarten, C.M. Schneider, and J. Kirschner, Phys. Rev. B **44**, 4406 (1991).
[19] D.G. Van Campen, R.J. Pouliot, and L.E. Klebanoff, Phys. Rev. B **48**, 17 533 (1993).
[20] Without the Co buffer layer a 12-ML Ni film grown directly on Cu(100) would have its easy axis of magnetization along the surface normal.
[21] A.K. See and L.E. Klebanoff, Phys. Rev. Lett. **74**, 1454 (1995); Phys. Rev. B **51**, 11 002 (1995).
[22] G. van der Laan, Phys. Rev. B **51**, 240 (1995)
[23] D. Venus, L. Baumgarten, C.M. Schneider, C. Boeglin, and J. Kirschner, J. Phys. Condens. Matter **5**, 1239 (1993).
[24] T. Jo and G.A. Sawatzky, Phys. Rev. B **43**, 8771 (1991).
[25] J.G. Menchero, Phys. Rev. Lett. **76**, 3208 (1996).
[26] L.E. Klebanoff, D.G. Van Campen, and R.J. Pouliot, Phys. Rev. B **49**, 2047 (1994).
[27] S. Raaen, Solid State Commun. **60**, 991 (1986).
[28] J. Cerdá, P. de Andres, A. Cebollada, R. Miranda, E. Navas, P. Schuster, C.M. Schneider, and J. Kirschner, J. Phys. Condens. Matter **5**, 2055 (1993).
[29] C.M. Schneider, P. Schuster, M. Hammond, H. Ebert, J. Noffke, and J. Kirschner, J. Phys. Condens. Matter **3**, 4349 (1991).