RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 44, NUMBER 21 1 DECEMBER 1991-I

Observation of magnetic circular dichroism in uv photoemission from ferromagnetic fcc cobalt films

C. M. Schneider, M. S. Hammond, and P. Schuster
Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-1000 Berlin 33, Germany

A. Cebollada and R. Miranda
Departamento de Física de la Materia Condensada, Universidad Autónoma de Madrid, Cantoblanco, E-28006 Madrid, Spain

J. Kirschner
Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-1000 Berlin 33, Germany

(Received 29 July 1991)

We report the observation of effects due to spin-orbit interaction in angle-resolved photoemission from an itinerant ferromagnet (fcc cobalt) using circularly polarized vuv synchrotron radiation. The photoelectron spectra of the Co 3*d*-band region are found to depend on the relative orientation of the sample magnetization and the photon spin. This effect may be understood as a form of magnetic circular dichroism appearing in the uv photoemission regime. It may be used to investigate magnetic phenomena in ferromagnets without performing an explicit spin-polarization analysis.

Apart from their well-known relevance in atomic levels, effects due to spin-orbit coupling are also found in the valence electronic states. These effects, although often involving small energy corrections, are crucial because they lift degeneracies of the electronic bands at high-symmetry points or lines in k space.[1] Spin-orbit coupling also breaks the rotational symmetry of the solid in the spin space, leading to a spin Hamiltonian with anisotropic coupling, i.e., the exchange coupling between neighboring spins in the solid depends on their spatial orientation relative to the crystal axes.[2] As a consequence, a single-crystal ferromagnet will be preferentially magnetized along certain crystallographic directions, as was demonstrated more than 60 years ago by Honda and Kaya.[3] These magnetocrystalline anisotropies, which play an important role in technology, indirectly confirm the presence of spin-orbit coupling in the valence electronic states of an itinerant ferromagnet.

Despite their importance, there is still very little direct experimental information about the changes in the electronic structure of a 3*d* ferromagnet introduced by spin-orbit interaction. Even in photoelectron spectroscopy, usually the method of choice to investigate band structures, this aspect of the electronic structure has been mostly neglected. This is somewhat surprising, since relativistic effects in paramagnets of comparable nuclear-charge number Z (e.g., copper) have been clearly identified in spin- and momentum-resolved photoemission experiments using circularly polarized light.[4] In paramagnets, the use of circularly polarized light results in spin-dependent photoexcitation matrix elements. This in turn yields photoelectrons of a distinct spin character, when they are derived from a spin-orbit split band. The photoelectron spin quantization axis is defined by the photon spin and the crystalline symmetry, which explains why this process has been termed "optical orientation".[5] By analogy, similar experiments performed on ferromagnets might yield information about the spin-orbit coupling in spin-polarized systems.

Recently, Baumgarten *et al.*[6] reported a form of magnetic circular dichroism (MCD) which appears in core-level photoemission from ferromagnetic iron. The authors observed that both the intensity and the line shape of the Fe 2*p* photoemission features depend on the relative orientation of the sample magnetization and the spin of the incident circularly polarized photons. This behavior is attributed to an exchange interaction in the core levels which further splits $2p_{1/2}$ and $2p_{3/2}$ states (already split by spin-orbit coupling) into spin-polarized sublevels with definite azimuthal quantum number m_J and energy eigenvalue $E(m_J)$.[7] The excitation probabilities of these sublevels depend on their spin character and the helicity of the incident light. This leads to a distinct difference in the intensity and the energy position of the spectral features, when comparing the spectra recorded with left- and right-hand circularly polarized light, respectively. This effect has been termed MCD in photoemission. The reported MCD in core-level spectroscopy thus involves a strong spin-orbit interaction ($\Delta E_{so} \sim 10$ eV) in combination with a weak exchange interaction ($\Delta E_{xc} \sim 0.5$ eV). This raises the question, whether a similar mechanism might exist in the itinerant electronic states of a 3*d* ferromagnet, where $\Delta E_{so} \ll \Delta E_{xc}$ is expected.

This question has been addressed with photoemission experiments performed at the German storage ring BESSY using the 6.5-m Normal Incidence Monochromator (NIM). This device offers circularly polarized radiation from the visible up to 35-eV photon energy with a degree of circularity of about 90%. Details of the experimental setup[4] and the 6.5-m NIM (Ref. 8) may be found elsewhere. The fcc cobalt samples consisted of ultrathin films of typically five monolayers thickness grown epitaxi-

ally on carefully prepared and characterized Cu(001) surfaces.[9] Previous studies indicated a good agreement between the films' band structure and three-dimensional fully relativistic calculations.[10] The films have been remanently magnetized along a ⟨110⟩ in-plane easy axis,[11] which has been oriented parallel to the scattering plane, as defined by the wave vector of the incident light **q** and the surface normal **n**. The angle of light incidence, θ, within the scattering plane is measured relative to the surface normal. To correlate our data with recent band mapping results along the [001] direction,[10] only normally emitted electrons were analyzed, thus probing the ΓX line in the bulk Brillouin zone.

The experiments described below have been performed for three different directions of the impinging light (see insets, Fig. 1). In the most symmetric arrangement, the light is normally incident. Therefore, the electric-field vector of the exciting radiation is always within the surface plane and the photon spin is perpendicular to the magnetization **M**. In the second geometry oblique incidence in the ⟨110⟩ mirror plane has been chosen (with the angle of incidence being $\theta=64°$ or $-64°$). The electric field thus contains a significant component perpendicular to the surface, and additional electronic transitions

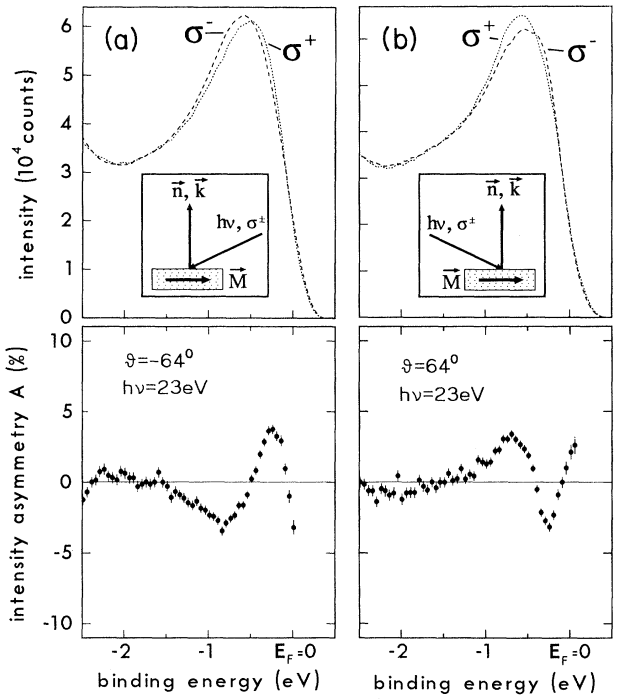

FIG. 1. (a) Magnetic circular dichroism in photoemission with circularly polarized light of $h\nu=23$ eV photon energy from fcc cobalt. Upper panel: Spin-integrated EDCs of the Co 3*d*-bands recorded at normal emission with σ^+ (dotted line) and σ^- light (dashed line), the angle of incidence denoted θ. Inset: Experimental geometry within the scattering plane giving the orientation of the remanent sample magnetization **M** relative to the surface normal **n** and the incident photon beam of helicity $\sigma^{\pm}$. Bottom panel: Corresponding intensity asymmetry distribution A as defined in the text. (b) Same as (a), but angle of incidence $\theta=64°$.

are allowed.[12] At the same time, the photon spin becomes partly aligned with the magnetization **M**.

Experiments performed in the normal emission and normal incidence geometry yield the following results. The energy distribution curves (EDCs) of the cobalt 3*d*-band region are virtually identical for both helicities of the circularly polarized light. Consequently, this particular geometrical arrangement, with the sample magnetization being *perpendicular* to the photon spin, does not give rise to a MCD signal outside the statistical uncertainty. The situation becomes completely different, however, if the experiment is performed with obliquely incident light. Now the energy distribution curve changes as a function of the helicity of the incident light and the sample magnetization. EDCs in this geometry have been measured at several photon energies for both positive (σ^+) and negative (σ^-) helicity. Representative spectra obtained at $h\nu=23$ eV are displayed in Fig. 1(a). The upper panel gives the EDCs for both helicities separately. The emission between E_F and -2 eV binding energy is predominantly due to direct interband transitions in fcc Co involving initial states of $\Delta_1^{\downarrow}$ and $\Delta_5^{\downarrow}$ single group symmetry character.[10] The two curves in Fig. 1(a) differ distinctly regarding the intensity and the energetic position of the dominant spectral feature. The intensity asymmetry distribution $A=[I(\sigma^+)-I(\sigma^-)]/[I(\sigma^+)+I(\sigma^-)]$ is a quantitative measure of this difference. $I(\sigma^+)$ and $I(\sigma^-)$ denote the intensity of the individual EDCs recorded with circularly polarized light of positive and negative helicity, respectively. This asymmetry is displayed on the lower panel of Fig. 1(a). It exhibits a pronounced structure in the energy region of the dominant peak, the maximum value of A being about 4% (not corrected for the incomplete polarization of the incident light).

Figure 1(a) clearly demonstrates that the energy distribution curve for a given geometry depends on the helicity of the exciting radiation. This result suggests a coupling between the spins of the photon and the excited photoelectron, as occurs in the presence of spin-orbit interaction. This effect may be understood as a kind of MCD in valence-band photoemission, and is thus the vuv counterpart to the observations reported in the soft-x-ray regime.[6] This interpretation receives further support from the finding that the asymmetry A depends on the *relative* orientation between the magnetization **M** and the spin of the incident photon. Within our experimental accuracy, the asymmetry A (with respect to the helicity) reverses if the sample is magnetized in the opposite direction. It also reverses if the direction of light incidence is changed to the opposite side of the vertical mirror plane. The latter situation is illustrated in Fig. 1(b), where the spectra measured at $\theta=64°$ are shown. Experimentally, the operations $\mathbf{M}\rightarrow-\mathbf{M}$ and $\theta\rightarrow-\theta$ turn out to be equivalent with respect to the intensity asymmetry A.

The above results can only be explained by invoking the presence of spin-orbit coupling which introduces a spin dependence into the excitation matrix elements. The reason for this becomes clear by considering the photoemission process in the nonrelativistic limit. The initial state in ferromagnets belongs either to a minority ($|\downarrow\rangle$) or majority ($|\uparrow\rangle$) band. The spin character is preserved dur-

ing the optical excitation and the emitted photoelectrons are thus spin polarized with a vector **P** which is always oriented parallel ($|\downarrow\rangle$) or antiparallel ($|\uparrow\rangle$) to the remanent magnetization **M**. Since the photon field cannot interact with the electron spin in the absence of spin-orbit coupling, the transition matrix elements are independent of the electron spin and excitation with σ^+ and σ^- light will yield identical EDCs. This is evidently in contrast to the observed intensity asymmetries in Fig. 1.

The inclusion of spin-orbit coupling introduces some new aspects. First, most of the band degeneracies will be lifted. Thus the overall number of energetically distinct bands increases [along a (001) direction in a cubic lattice only Δ_5 states split[13]] and bands with different single group symmetry may hybridize. In some regions of the Brillouin zone this process will even mix bands of opposite spin character.[14] In addition, the electron spin will be coupled to the orbital part of the electron wave function and the transition matrix elements become spin dependent. Therefore, a mechanism similar to the optical orientation in paramagnets should become possible in ferromagnets, too.

In contrast to a paramagnet, each initial state in a ferromagnet is split into distinct energy eigenvalues for the spin-down ($\downarrow$) and spin-up ($\uparrow$) electrons. The mean value of this so-called exchange splitting in fcc Co is $\Delta E_{xc} \approx 1.2$ eV for the Δ_5 bands. The quantization axis of the exchange split states is defined by the magnetization **M** within a ferromagnetic domain. Recalling from above, some of these bands may still be degenerate due to their spatial (nonrelativistic) symmetry. Spin-orbit interaction lifts this degeneracy and causes a further splitting of the states of the order of $\Delta E_{so} \approx 0.1$ eV, i.e., $\Delta E_{so} \ll \Delta E_{xc}$ in fcc Co. The spin-orbit split bands have different photoexcitation probabilities due to their relativistic symmetry character. This results in different energy distribution curves upon the excitation with light of positive and negative helicity, and hence an intensity asymmetry as has been observed in the experiments. It should be noted that this form of intensity asymmetry cannot occur in relativistic paramagnets, since the initial states are not spin polarized, i.e., the energy eigenvalue does not depend on spin. The difference in the excitation with left and right circularly polarized light therefore shows up in the *spin polarization* of the excited photoelectrons, but not in the energy-distribution curve as is the case in ferromagnets.

In the frame of the three-step model of photoemission, magnetic circular dichroism in uv photoemission may be interpreted as the interference between an initial state effect (spin-polarized state) and an operator effect (spin-dependent dipole matrix elements) and the corresponding spin quantization axes. The quantization axis of the electrons in the initial state is defined by the magnetization **M** (oriented along a ⟨110⟩ direction within the film plane). In the excitation step, however, the incident light introduces a new quantization axis **Q** due to the photon spin. In the particular case of normal incidence, i.e, $\mathbf{M} \perp \mathbf{Q}$ in the present system, a spin-dependent excitation will not occur, since the expectation value of the electron spin along **Q** is zero. This is in agreement with the experimental results at normal incidence, where **M**—and thus the electron spin—is orthogonal to the photon spin and the intensity asymmetry vanishes. To obtain a nonzero MCD signal therefore requires the photon spin to be partially aligned with the magnetization **M**. This qualitative interpretation of the observed magnetic circular dichroism in the valence bands of an itinerant ferromagnet should be substantiated by photocurrent calculations, which must treat exchange and spin-orbit interaction on an equal footing. Such theoretical investigations of the valence electronic states are, to our knowledge, not available at present.

In conclusion, we have demonstrated the importance of relativistic interactions in the band structure of ferromagnets. The influence of spin-orbit coupling gives rise to a novel effect, which may be interpreted as magnetic circular dichroism in valence band photoemission. Due to its inherent surface sensitivity, this effect could be a useful tool in future experiments on surface magnetism and on the role of spin-orbit interaction in ferromagnets. Because MCD involves only intensity measurements, it offers a more elegant and faster way to determine the spin of the excited photoelectrons from ferromagnets without the need of an explicit spin polarization analysis. MCD in valence-band photoemission might be used to image ferromagnetic domains in real time. For this purpose, the photoelectrons may be excited just above the threshold and used to analyze the surface by means of a photoemission microscope.[15] Since the MCD signal (intensity asymmetry) depends on the relative orientation of the magnetization and the photon spin, photoemission from domains which are magnetized parallel and antiparallel to the photon spin will give rise to a photoemission yield which varies spatially across the sample surface. This spatial intensity distribution can be translated into a black and white contrast to display an image of the domain pattern on a television screen. As an advantage over scanning methods like scanning electron microscopy with polarization analysis[16–18] or Lorentz microscopy,[19] this technique will permit the investigation of magnetization dynamics at ferromagnetic surfaces.

We thank D. Venus for fruitful discussions and a critical reading of the manuscript. Thanks are also due to E. Navas for his help during the experiments. This work was supported by Bundesministerium für Forschung und Technologie under Contract No. 05 413 AXI, TP 3.

[1] J. C. Slater, *Quantum Theory of Molecules and Solids II* (McGraw-Hill, New York, 1965).

[2] R. M. White, *Quantum Theory of Magnetism* (Springer-Verlag, Berlin, 1983).

[3] K. Honda and S. Kaya, Sci. Rep. Tohoku Univ. **15**, 721 (1926); S. Kaya, *ibid.* **17**, 639 (1928); **17**, 1157 (1928).

[4] C. M. Schneider, J. J. de Miguel, P. Bressler, P. Schuster, R. Miranda, and J. Kirschner, J. Electron Spectrosc. Relat.

Phenom. **51**, 263 (1990).

[5]Wöhlecke and G. Borstel, in *Optical Orientation,* edited by F. Meier and B. P. Zakharchenya (North-Holland, Amsterdam, 1984).

[6]L. Baumgarten, C. M. Schneider, H. Petersen, F. Schäfers, and J. Kirschner, Phys. Rev. Lett. **65**, 492 (1990).

[7]H. Ebert, J. Phys. Condens. Matter **1**, 9111 (1989); H. Ebert, L. Baumgarten, C. M. Schneider, and J. Kirschner, Phys. Rev. B **44**, 4406 (1991).

[8]F. Schäfers, W. Peatman, A. Eyers, C. Heckenkamp, G. Schöhense, and U. Heinzmann, Rev. Sci. Instrum. **57**, 1032 (1985).

[9]C. M. Schneider, J. J. de Miguel, P. Bressler, P. Schuster, R. Miranda, and J. Kirschner, Phys. Rev. Lett. **64**, 1059 (1990).

[10]C. M. Schneider, P. Schuster, M. S. Hammond, H. Ebert, J. Noffke, and J. Kirschner, J. Phys. Condens. Matter **3**, 4349 (1991).

[11]H. P. Oepen, M. Benning, H. Ibach, C. M. Schneider, and J. Kirschner, J. Magn. Magn. Mater. **86**, L137 (1990).

[12]J. Hermanson, Solid State Commun. **22**, 9 (1977).

[13]S. C. Miller and W. F. Love, *Tables of Irreducible Representations of Space Groups and Co-representations of Magnetic Space Groups* (Pruett, Boulder, 1967).

[14]B. Ackermann, R. Feder, and E. Tamura, J. Phys. F **14**, L173 (1984); L. Fritsche, J. Noffke, and H. Eckardt, J. Phys. F **17**, 943 (1987).

[15]G. Mollendstedt and F. Lenz, Adv. Electron. Electron Phys. **18**, 251 (1963); W. Telieps and E. Bauer, Surf. Sci. **162**, 163 (1985).

[16]K. Koike and K. Hayakawa, J. Appl. Phys. **23**, L187 (1984).

[17]J. Unguris, G. Hembrel, R. J. Celotta, and D. T. Pierce, J. Microsc. (Oxford) **139**, RP1 (1985).

[18]H. P. Oepen and J. Kirschner, Phys. Rev. Lett. **62**, 819 (1989).

[19]J. N. Chapman and G. R. Morrison, J. Magn. Magn. Mater. **35**, 254 (1983).