Magnetic dichroism in angle-resolved UV photoemission from valence bands, using linearly polarized light

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Magnetic dichroism measurements of the valence bands of films of fcc Co/Cu (001) have been performed using angle-resolved UV photoemission in low-symmetry, off-normal emission geometries, and linearly polarized light. Asymmetries of magnitude 4% are observed upon magnetization reversal. Evidence of both spin-dependent surface transmission and magnetic dichroism in the angular distribution of photoelectrons is seen. The asymmetry spectra indicate sensitivity to both magnetic exchange splitting and to spin–orbit splitting. © *1996 American Institute of Physics.* [S0021-8979(96)78808-3]

Magnetic dichroism in the angular distribution of photoelectrons (MDAD) makes it possible to study the interplay between the magnetic exchange and spin–orbit interactions in solids. To date, it is primarily the deep[1,2] and shallow core[3,4] levels of magnetic materials which have been investigated in this way. Over the course of these studies, a series of findings have overturned expectations, and lead to a better understanding of the technique and its potential. It turns out that the dichroism is sizable not only in the deepest core levels, that circularly polarized light is not required, and a larger dichroism may often be obtained using linearly polarized light;[3,5] and that the dichroism from core levels is not independent of emission angle, but contains significant crystallographic information.[1,6] These findings all suggest the application of MDAD to valence band studies, to see what can be learned about the magnetically important states near the Fermi level without recourse to more complicated spin-resolved experiments, or to monochromators specialized for the production of circularly polarized light. All that is required is a conventional angle-resolved photoemission apparatus, a source of linearly or unpolarized monochromatic light, and a means to reverse the remanent magnetization of the sample.

A few experimental studies which confirm the existence of MDAD in the valence bands have been reported, but most have used circularly polarized light,[7] and all have used normal emission geometries which restrict access to a few regions of the Brillouin zone.[8] Furthermore, a straightforward qualitative model giving an overview of the mechanism by which the dichroism arises in valence band emission is still missing. It is therefore difficult to interpret the measurements without recourse to specialized one-step photoemission calculations with nonperturbative treatments of the exchange and spin–orbit coupling in the surface electronic band structure.[9] These calculations are not yet available for general, low symmetry experimental geometries. The purpose of the present experiments is, in the first place, to observe the magnetic dichroism in the valence bands in off-normal emission geometries, using linearly polarized light. With measurements in hand, it may be possible to better assess how the dichroism can be used in a practical sense to learn about the electronic structure at magnetic surfaces.

An example of the ambiguities which arise in magnetic dichroism experiments in the valence bands, is the question of the conceptual origin of the observed dichroism. For concreteness, consider the experimental geometry in Fig. 1. The remanent magnetization, **M**, is in the surface of the sample crystal, and both the incident light wave vector, **q**, and the photoelectron wave vector, **k**, lie in the *xy* plane normal to **M**. The light is linearly polarized in this plane. In Fig. 1, **q** and **k** are collinear, but this need not be the case. It is further assumed that the *xz* plane containing **M** is a mirror symmetry plane of the crystal—a common situation among itinerent ferromagnets.

The experiment involves collecting two angle-resolved photoelectron energy distributions—one for each of the two reversed remanently magnetized states of the sample. The difference in these two energy distributions is the energy- and angle-resolved dichroism. There are two mechanisms which may contribute to the dichroism. The first is spin-dependent transmission.[10] It is most apparent as a ''final state effect'' in a three-step model of photoemission, where photoexcitation occurs between bulklike electronic states and the photoelectrons are subsequently transmitted to the vacuum. At the surface, the photoelectron states in the crystal (which are not pure spin states, because of spin–orbit coupling) must be matched to those in the vacuum (which are pure

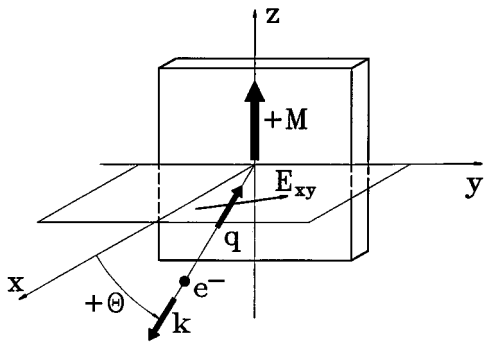

FIG. 1. The Co(001) film has remanent magnetization along $\pm z$, and lies in the *xz* mirror plane. The incoming light with wave vector **q** (with linear polarization $\mathbf{E}_{xy}$) and the photoelectron wave vector **k** all lie in the *xy* mirror plane, making an angle θ with the *x* axis.

 0021-8979/96/79(8)/6504/3/$10.00

spin states). This leads to spin-dependent transmission coefficients. In the above experiment, reversing the magnetization reverses the spin character of the photoelectrons, and the altered transmission coefficient may cause the dichroism. Recent results for Cu show that this mechanism can cause an asymmetry of $\pm 3\%$ in $3d$ metals.[11]

MDAD is typically considered to arise from the photoexcitation step itself.[12,13] The spectra taken with reversed magnetization compare two inequivalent experimental geometries which are related by a mirror reflection—a traditional statement of dichroism. This can be understood with reference to Fig. 1, where reversal of the magnetization is equivalent to reflection in the *xz* mirror plane of the crystal. However, this operation also alters the experimental quantities **k**, **q**, and $\mathbf{E_{xy}}$. In particular, the even and odd parts of $\mathbf{E_{xy}}$ and the photoelectron wave function undergo a relative change in phase, and the resulting photoexcitation matrix element will have both even and odd parts. Upon squaring and subtracting to form the dichroism, only the interference terms between the even and odd parts of the matrix element survive, giving the dichroism.[13] Because of its origin in the transition matrix elements, MDAD is usually classified as an ''initial state effect.''

The view that magnetic dichroism arises from MDAD when it is an initial state effect and from spin-dependent transmission when it is a final state effect cannot be considered as a rigid classification. However, alternative classifications are worse: In the three-step model of photoemission, spin-dependent transmission exists, but MDAD (with linearly polarized light) does not. In a one-step model, all the dichroism must be assigned to the matrix elements—that is MDAD. These classifications are not as useful for qualitative arguments, and ultimately for the understanding of experiments.

The photoemission experiments were carried out at the BESSY synchrotron storage ring, using a 6.5 normal incidence monochromator, and an angle-resolved photoemission apparatus which is described elsewhere.[14] The samples were 5 or 6 monolayer fcc Co films grown on a Cu(001) substrate. The film growth was monitored using medium energy electron diffraction.[15] Hysteresis loops measured by the magneto-optic Kerr effect confirmed that the films were remanently magnetized in the plane of the surface. The electron energy analyzer had a hole in the back, which allowed the light beam to pass through the entrance lenses, and permitted the experimental geometry with $\mathbf{k}=-\mathbf{q}$, as shown in Fig. 1. This geometry has three important attributes: (a) A wide range of off-normal emission conditions can be reached by a single rotation of the sample about the **z** axis. (b) Since **k**, **q**, and **M** are coplanar, oriented atom models of MDAD predict no dichroism[12]—any observed effects will be related to crystallographic information. (c) The *xy* plane is a true mirror plane for the magnetic system plus the experiment. This latter point implies that MDAD using either linearly or circularly polarized light measures the same matrix elements, as has been confirmed theoretically[13] and experimentally.[16] A disadvantage of this geometry is that the light passing through the spectrometer creates more secondary electrons than is usual, and this limits the useful energy range of the spectra, particularly at lower photon energies.

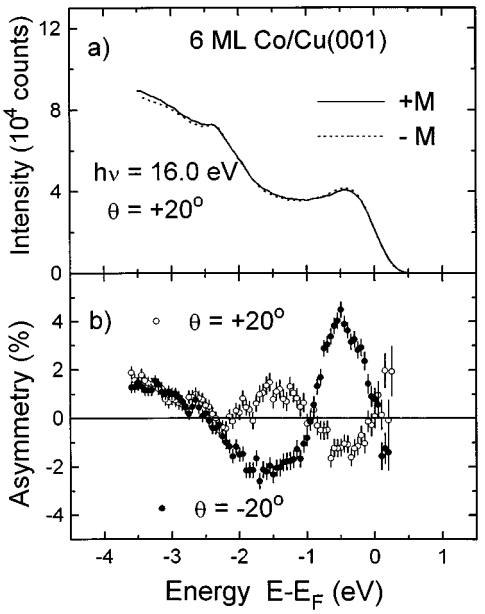

FIG. 2. (a) Photoelectron intensity spectra for two senses of magnetization, with a photon energy of 16.0 eV and emission angle $+20°$. (b) The resulting intensity asymmetry for the data in part (a) (open symbols), and for data measured at an emission angle of $-20°$ (closed symbol).

Experimental results for a photon energy of 16.0 eV, with $\theta=+20°$ are shown in Fig. 2. Part (a) presents the separate intensity spectra for positive and negative magnetization. The spectra have had a linear background (determined before the Fermi energy) removed, and have been normalized to compensate for changes in the photon flux during the course of the measurements. The open symbols in part (b) show the intensity asymmetry, defined as the difference of the two spectra divided by their sum. It shows clear positive and negative peaks in the energy range down to about -2.5 eV where the Co *d* bands lie, and a gradual upward slope at lower energy. The asymmetry of a similar pair of data sets collected at $\theta=-20°$ are also plotted in Fig. 2(b). The asymmetry peaks for this angle are reversed in the energy region of the Co bands, as they must be if they are due to either MDAD or spin-dependent transmission. The slope in the region of secondary electrons depends instead on the absolute sign of the magnetization, and probably represents a perturbation of the secondary electrons generated by the light passing through the spectrometer.

It can be seen immediately that the magnitude of the dichroism in off-normal emission is in the range of 2%–4% when secondary electrons are not removed. This is the same approximate size as that seen in previous experiments in normal emission, and is also the same as typical (rather than maximum) values seen in MDAD from core levels when the secondary electrons are not removed. MDAD experiments in the valence bands are not significantly more difficult than those in the core levels. This example also demonstrates the advantage of using a differential spectroscopy such as MDAD—it is possible to display clearly the presence of Co states near -1.5 eV despite the fact that they do not appear

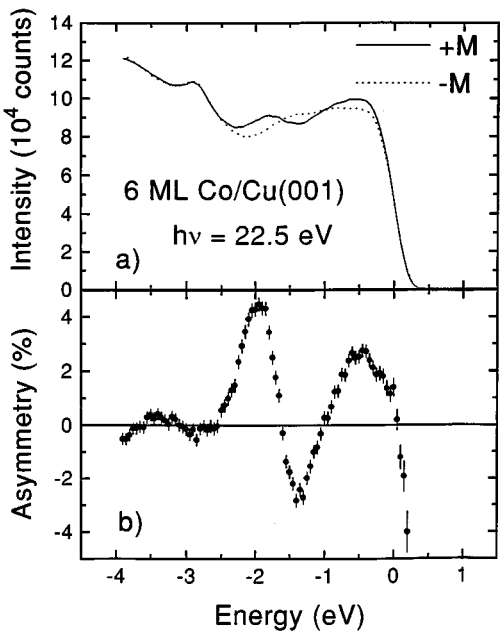

FIG. 3. (a) Photoelectron intensity spectra for two senses of magnetization, with a photon energy of 22.5 eV and emission angle −16°. (b) The resulting intensity asymmetry for the data in part (a).

as peaks in the intensity spectra. The form of the dichroism spectra suggests that, in this instance, the dichroism arises primarily due to spin-dependent transmission at the surface. The first-order perturbation observed in the intensity spectra is a change in the size of the peak—there is no indication that the intensity peak near −0.5 eV is shifted when the magnetization is reversed. This behavior is expected for the final state effect of spin-dependent transmission upon spin reversal. The positive and negative peaks in the asymmetry are then interpreted as being due to separate transitions from states of opposite spin, which are separated by approximately 1.2 eV, in agreement with the exchange splitting found in calculations and spin-resolved photoemission experiments[17] for fcc Co. Note that the absolute sign of the asymmetry is not easily related to the spin of the photoelectrons, since it depends on the surface transmission coefficients.

The data presented in Fig. 3 for a photon energy of 22.5 eV and $\theta = -16°$ show a different behavior. The peak below about −2.5 eV is due to the Cu substrate, and shows no asymmetry. However, a well-resolved peak at −1.8 eV seen with $+\mathbf{M}$ shifts to −1.6 eV when the magnetization is reversed, but the peak intensity is not greatly affected. This is the behavior expected for MDAD. The first-order effect of weak spin–orbit coupling on the initial states is to introduce hybridization between nearly degenerate bands of opposite spin. While the energy of the hybridized states depends on the relative orientation[18] of the spin σ and $\mathbf{M}$, as $\sigma+\mathbf{M}$, the transition matrix elements depend as well on the absolute spin direction through the spin–orbit coupling $\sigma \cdot l$. Thus the transition with a given orientation of $\sigma \cdot l$ occurs at a different energy when $\mathbf{M}$ is reversed, leading to the apparent shift in the peak energy in the intensity spectra. A rapid minus/plus variation in the asymmetry results. The experimentally observed energy shift is of the magnitude expected for the spin–orbit interaction for $3d$ metals. These spectra coincidentally demonstrate that MDAD persists in this low-symmetry geometry, where $\mathbf{q}$, $\mathbf{k}$, and $\mathbf{M}$ are coplanar, even though models based on photoemission from oriented atoms predict a null effect. This is a consequence of the crystalline symmetry, rather than atomic symmetry, of the system, and is not surprising for such low photon energies.

The asymmetry close to the Fermi level in Fig. 3 is not as clear cut as the previous two examples, and more a detailed analysis is called for. It is not yet clear whether or not a simple criterion can be found to distinguish MDAD from surface transmission effects. Analysis of the systematics of a more comprehensive data set in underway to determine how widely the present classification of asymmetry features is applicable.

[1] C. M. Schneider, D. Venus, and J. Kirschner, Phys. Rev. B **45**, 5041 (1992).
[2] G. D. Waddill, J. G. Tobin, and D. P. Pappas, Phys. Rev. B **45**, 552 (1992).
[3] Ch. Roth, F. U. Hillebrecht, H. B. Rose, and E. Kisker, Solid State Commun. **86**, 673 (1993).
[4] G. Rossi, F. Sirotti, N. A. Cherepkov, F. Combet-Fornoux, and G. Panacionne, Solid State Commun. **90**, 557 (1994).
[5] D. Venus, Phys. Rev. B **48**, 6144 (1993).
[6] D. Venus, L. Baumgarten, C. M. Schneider, C. Boeglin, and J. Kirschner, J. Phys. Condens. Matt. **5**, 1239 (1993).
[7] C. M. Schneider, M. S. Hammond, P. Schuster, A. Cebollada, R. Miranda, and J. Kirschner, Phys. Rev. B **44**, 12 066 (1991); J. Bansmann, C. Westphal, M. Getzlaff, F. Fegel, and G. Schönhense, J. Magn. Magn. Mater. **104–107**, 1691 (1992).
[8] M. Getzlaff, Ch. Ostertag, F. H. Fecher, N. A. Cherepkov, and G. Schönhense, Phys. Rev. Lett. **73**, 3030 (1994).
[9] N. A. Scheunemann, S. V. Halilov, J. Henk, and R. Feder, Solid State Commun. **91**, 487 (1994).
[10] H.-P. Oepen, K. Hünlich, and J. Kirschner, Phys. Rev. Lett. **56**, 496 (1986); J. Garbe and J. Kirschner, Phys. Rev. B **39**, 9859 (1989).
[11] D. Venus, W. Kuch, A. Dittschar, M. Zharnikov, C. M. Schneider, and J. Kirschner, Phys. Rev. B **52**, 6174 (1995).
[12] B. J. Thole and G. van der Laan, Phys. Rev. B **49**, 9613 (1994).
[13] D. Venus, Phys. Rev. B **49**, 8821 (1994).
[14] H. P. Oepen, Ph.D thesis, Rhenische-Westfälische Technische Hochschule, Aachen, Germany, 1984.
[15] C. M. Schneider, J. J. de Miguel, P. Bressler, J. Garbe, S. Ferrer, R. Miranda, and J. Kirschner, J. Phys. C **8**, 1657 (1988).
[16] F. U. Hillebrecht, Ch. Roth, H. B. Rose, M. Finazzi, and L. Braicovich, Phys. Rev. B **51**, 9333 (1995).
[17] C. M. Schneider, P. Schuster, M. Hammond, H. Ebert, J. Noffke, and J. Kirschner, J. Phys. Condens. Matter **3**, 4349 (1991).
[18] It is assumed that the initial state is not a surface state.