Angle-resolved study of magnetic dichroism in photoemission using linearly polarized light

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The angular dependence of the magnetic dichroism in core-level photoemission using linearly polarized light has been measured. The intensity asymmetry of Co 3*p* spectra of Co/Cu(100) ultrathin films upon magnetization reversal has been determined in normal emission, under variation of both polar and azimuthal angles of light incidence. The results are compared to general theoretical predictions of angle-resolved x-ray magnetic dichroism for photoemission along directions of high symmetry. Good agreement between the experimental results and the predicted angular dependences is found, thus confirming that interference terms in the dipole matrix elements are responsible for the angle-resolved magnetic dichroism.

Magnetic dichroism in the angular distribution of photoelectrons (MDAD) has proven to be a versatile technique for the investigation of magnetic surfaces and thin films. It may be used to characterize magnetic systems qualitatively, and to probe the interplay between the exchange potential and the spin-orbit coupling which is at the basis of magnetism. Magnetic dichroism investigating core levels was performed in angle-integrated absorption experiments,[1] and interpreted with angle-integrated theories of photoemission from isolated, oriented atoms.[2] Angle-resolved magnetic dichroism in photoemission experiments are a more recent development,[3] and a comprehensive understanding of MDAD is emerging only now. Experiments at the Fe 2*p* levels with circularly polarized light[4,5] showed that MDAD upon magnetization reversal has a strong angular dependence upon the light wave vector **q**, the photoelectron wave vector **k**, the magnetization **M**, and the surface normal **n**. This was interpreted qualitatively using a one-electron photoemission theory. The important role played by interference terms in the dipole photoexcitation matrix elements was soon realized,[6,7] and lead to the suggestion that MDAD should also be observed with linearly, and with unpolarized light.[7] This was experimentally verified independently by Roth *et al.*[8] for linearly polarized light. Three subsequent analyses[9–11] have derived relations between various angle-resolved magnetic dichroism experiments, and indicated which photoexcitation matrix elements are probed by each. They conclude that, for emission geometries of high symmetry, the angular dependence of MDAD upon magnetization reversal is decoupled from the shape of the spectrum. Furthermore, the shape of the spectrum is independent of the light polarization. A straightforward method to test these recent theoretical interpretations is to measure the angular dependences of MDAD using linearly polarized light. At present, only a few fixed geometries have been investigated.[8,12] The purpose of this communication is to present experimental results for the systematic variation of MDAD with the vector quantity **q**.

MDAD using linearly polarized light can be thought of as an interference effect. The dipole photoexcitation matrix element must be the sum of both even and odd parts, so that even though the size of each part remains the same under reversal of the magnetization, the interference cross terms changes and gives an asymmetry. The theories mentioned above treat MDAD either in an atomic picture[10,11] or in one-electron photoemission from a solid.[9] The former are based on photoemission from an isolated, polarized atom with spherical symmetry, and consider photoexcitation from localized shells to a continuum of plane-wave states. The latter allows a more general final state that can include effects of the crystalline symmetry of the sample. All theories are identical for emission geometries of high symmetry (i.e., for normal electron emissions from a surface containing three or more reflection planes) since then only components of the final wave function of the form $Y_{l,0}(\mathbf{k})$ are coupled to the plane waves at the detector, and the crystalline and spherical environment cannot be distinguished. This is not true for emission geometries of lower symmetry, where the crystallinity of the sample has proven to be important.[5]

To derive the angular dependence of the dichroism for normal electron emission along a high symmetry direction, Eq. (22) of Ref. 9 may be used with $P_L=1$ and $P_C=0$. It yields for excitation with linearly polarized light with respect to magnetization reversal

$$D_L^M(\mathbf{q}, r=1, \alpha, \mathbf{k}=\mathbf{n}) = 4\sin\Theta\cos\Theta\cos\Phi\,\mathrm{Re}[\chi_{zy}]\,. \quad (1)$$

The angle of incidence Θ and the azimuthal angle Φ of the incident light are explained in Fig. 1, which shows the geometrical setup. The notation $r=1$ denotes linearly polarized light, and the polarization angle $\alpha(\Theta,\Phi)$ is such

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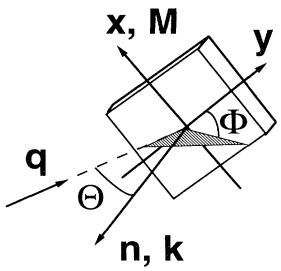

FIG. 1. Experimental geometry, showing the vectors of photon momentum **q**, electron momentum **k**, magnetization **M** and surface normal **n**. The light is incident under a polar angle Θ with respect to **n** with the linear polarization in the reaction plane defined by **q** and **n**. **x** and **y** are ⟨110⟩ symmetry directions along the sample surface. The magnetization **M** is either in **x** or in $-\mathbf{x}$ direction. Φ is the azimuthal angle by which **y** is tilted away from the reaction plane.

that the polarization axis is in the reaction plane defined by **q** and **n**. **x** and **y** are ⟨110⟩ symmetry directions along the sample surface. The magnetization **M** is changed from **x** to $-\mathbf{x}$ to display the dichroism. The matrix element χ_{zy} is given by the expression

$$\chi_{zy}=\langle\psi_i(\mathbf{M})|z|\psi_f(k)\rangle\langle\psi_f(k)|y|\psi_i(\mathbf{M})\rangle\ . \qquad (2)$$

It contains interference terms arising from components of the dipole operator in z ($=-\mathbf{n}$) and **y** directions. $\psi_i(\mathbf{M})$ and $\psi_f(\mathbf{k})$ are the initial and final-state wave functions. The explicit $\sin\Theta\cos\Theta\cos\Phi$ angular dependence in Eq. (1) results from the projection of the polarization vector onto the z and y axes of the crystal. The matrix element χ_{zy} is given with respect to these directions and does not exhibit an angular dependence, i.e., angular functions appear in front of a fixed matrix element. It is worth mentioning that for the case of circularly polarized light ($P_L=0$ and $P_C=1$) the asymmetry D_C^M is predicted to have the same matrix element χ_{zy} with only a different size and angular behavior, as is also the case for elliptically polarized light.[9]

The system Co/Cu(100) is well suited to test these predictions owing to its well known and extensively studied epitaxial layer-by-layer growth[13–15] and its straightforward magnetic behavior. It shows an in-plane easy axis of magnetization in the ⟨110⟩ azimuth, with the Curie temperature well above room temperature for film thicknesses higher than 2 ML.[13,14] Cobalt films were deposited on Cu(100) at room temperature from a high-purity cobalt rod heated by electron bombardment with a typical deposition rate of 2 ML/min. Film thickness and growth conditions were monitored during the evaporation by medium-energy electron diffraction (MEED). The MEED intensity curves of the (00)-beam showed the oscillations indicative of layer-by-layer growth already presented in Ref. 13, and allowed a thickness calibration with an accuracy of better than 0.1 ML. The overall pressure in the chamber did not rise above 2×10^{-8} Pa during the preparation of the films, and no impurities could be detected after deposition using Auger electron spectroscopy. All measurements presented in this paper refer to Co films of 6.0-ML thickness. The sample was remanently magnetized in-plane in the ⟨110⟩ azimuth in the course of a magneto-optic Kerr-effect (MOKE) experiment. All films showed the well-known easy-axis hysteresis loops.[13] To check the presence of the magnetization, MOKE measurements were performed between and after the photoemission measurements with reversed magnetization. The sample temperature during the magnetization procedure and during the measurement of the photoelectron spectra was 300 K.

Linearly polarized light was taken from the crossed undulator beamline (U2-FSGM) at the Berlin synchrotron radiation facility (BESSY) at $h\nu=125$ eV, using only the horizontal undulator. The entrance and exit slits of the monochromator were set to 200 and 300 μm, respectively, resulting in an overall energetic resolution of approximately 1 eV. The spectrometer has been described elsewhere.[16] It allows the detection of electrons emitted either near to the direction of the incoming light or at angles greater than 20° off the direction of light incidence.

Figure 2(a) shows a representative spectrum for normal photoelectron emission and the light incident under an angle $\Theta=40°$ to the surface normal. The azimuthal angle is $\Phi=0$, so that the magnetization **M** is perpendicular to the reaction plane either in **x** direction (broken line) or in $-\mathbf{x}$ direction (dotted line). The sum of both spectra is reproduced as continuous line. Panel (b) shows the asymmetry A defined by $A=[I(\uparrow)-I(\downarrow)]/[I(\uparrow)+I(\downarrow)]$. It exhibits a plus-minus feature, reaching from $+5\%$ to -2.5%. The shape of the spectrum appears to be similar to that observed with Fe in the same geometry but under a different angle Θ.[8]

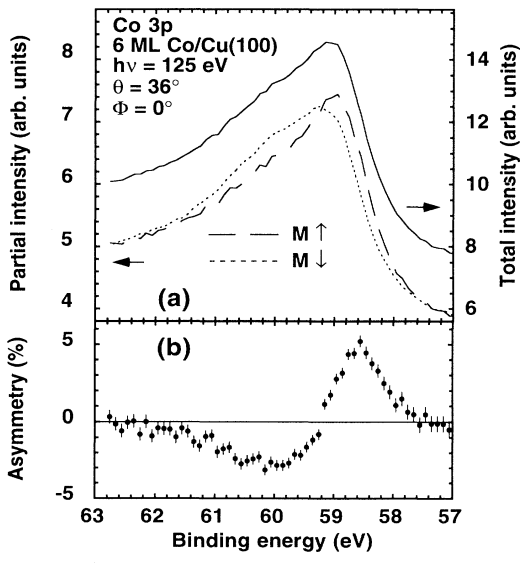

FIG. 2. (a) Co 3p photoelectron spectra of 6.0-ML Co/Cu(100) taken with linearly polarized light of 125-eV photon energy under an incidence angle of $\Theta=36°$ and an azimuthal angle $\Phi=0$ for the direction of film magnetization "up" ($\mathbf{M}=\mathbf{x}$, broken line) and "down" ($\mathbf{M}=-\mathbf{x}$, dotted line). The solid line reproduces the sum of both (b) Asymmetry A, calculated from the spectra of panel (a) by means of $A=[I(\uparrow)-I(\downarrow)]/[I(\uparrow)+I(\downarrow)]$.

One of the predictions of Eq. (1) is that the shape of the dichroism D_L^M does not depend on the emission geometry, since the angular dependence is decoupled and only affects the amplitude of the dichroism. Figure 3 shows a series of asymmetry spectra for different values of Θ with $\Phi=0$. It can be stated that the shape of the curves, a pronounced peak around 58.5-eV binding energy, and a smaller and broader feature of opposite sign around 60.1-eV binding energy, is the same for all spectra. Thus, varying Θ or Φ (not shown in Fig. 3) does indeed not affect the shape of the spectra, only their amplitude.

Figure 4 shows the angular dependences of the asymmetry A as a function of the angle of incidence Θ at fixed azimuthal angle $\Phi=0$ [Fig. 4(a)] and as a function of the azimuthal angle Φ at fixed polar angle $\Theta=36°$ [Fig. 4(b)], both at normal takeoff of the photoelectrons. In each case the sum over the height of the plus and minus extrema of the asymmetry spectra is shown. The vertical error bars refer to the uncertainty in the determination of A as estimated from the reproducibility of measurements from different days. They are somewhat larger than the pure statistical error [error bars in Fig. 2(b)]. A fit to the $\sin\Theta\cos\Theta$ law is given in Fig. 4(a) as solid line. Although Eq. (1) is valid for the difference D_L^M of spectra with opposite **M**, i.e., for the numerator of the asymmetry A, a comparison between D_L^M and A can be done because of the strong and smooth background superimposed on the spectra [note position of origin in Fig. 2(a)], which makes the denominator of the asymmetry essentially independent of angle. The solid line in Fig. 4(b) reproduces the $\cos\Phi$ dependence with an amplitude taken from the fit of Fig. 4(a). A good agreement between experiment and theory can be stated for both the variation of the polar angle Θ and the azimuthal angle Φ.

The experimental results prove directly the nature of the observed MDAD as a pure interference effect described by a product of matrix elements with dipole operators of different symmetry [Eq. (2)] and not by a modulus squared term. The effect can only exist in an angle-resolved experiment, since angle-integrated experiments integrate a product of odd and even functions to zero. By verifying the angular dependence given by Eq. (1), this fundamental aspect of the theories is confirmed. The $\sin\Theta\cos\Theta\cos\Phi$ dependence reflects the size of the component of the light polarization with respect to the mirror plane containing **M** and **n**. Interference of components of the dipole operator with different parity under reflection in that plane creates the observed MDAD. Having confirmed the theories by comparison with the angular dependence for linearly polarized light, we note that no change in the spectrum shape with light polarization is expected. Comparison of the present spectra to published data for $3p$ emission using circularly polarized light from Fe(110) (Ref. 5) and Ni(110) (Ref. 17) show that this is qualitatively correct.

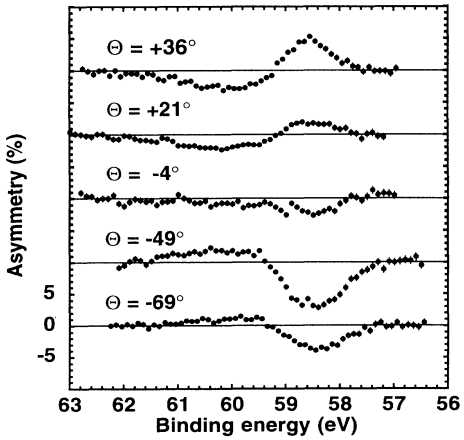

FIG. 3. Asymmetry spectra of Co $3p$ upon magnetization reversal for different angles Θ of light incidence with the azimuthal angle fixed at $\Phi=0$.

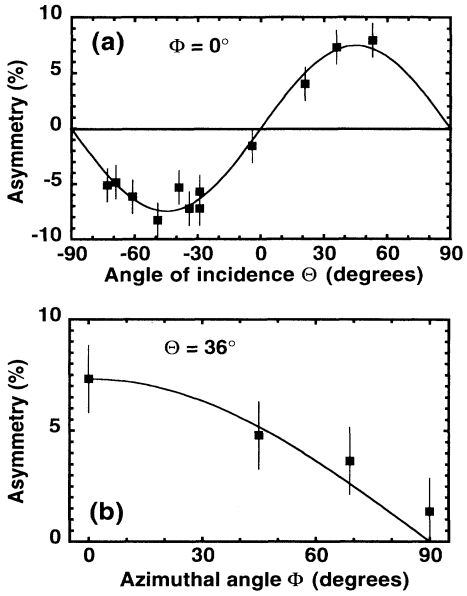

FIG. 4. Dependence of experimental dichroic asymmetry on angular variation. (a) Dependence on the polar angle of incidence Θ with fixed azimuthal angle $\Phi=0$. A fit to the $\sin\Theta\cos\Theta$ law is given as solid line. (b) Dependence on the azimuthal angle Φ with fixed polar angle $\Theta=36°$. The solid line represents a $\cos\Phi$ dependence, using as amplitude for $\Phi=0$, the value obtained from the fit in panel (a).

In conclusion, an experimental study of the angular dependence of MDAD using linearly polarized light was performed. To test recent theories on MDAD, the intensity asymmetry of Co $3p$ spectra under normal emission upon magnetization reversal was measured for Co/Cu(100) thin films. For variation of either the polar angle of incidence or the azimuthal angle of the magnetization direction, the theories were confirmed experimentally: the shape of the dichroic asymmetry does not depend on the emission geometry, and the angular variation of the amplitude of the dichroism follows a $\sin\Theta\cos\Theta\cos\Phi$ law, proving thus MDAD as a pure interference effect. It remains to be seen whether the more complicated angular dependence expected for emission geometries of lower symmetry[9] will be observed.

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