Use of Polarized Photons in Dichroism and Photoemission

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The application of polarized synchrotron radiation in surface-sensitive studies of magnetic materials is discussed. An overview of the various spectroscopies is given and applications of linearly- and circularly-polarized photons in x-ray absorption and photoemission are discussed.

I. Introduction

Synchrotron spectroscopies of magnetic materials are especially powerful when they take advantage of the ability to select the photon polarization. This selection of the photon polarization, whether linearly- or circularly-polarized allows one to set specific experimental geometries that provide much more information on magnetic material parameters than can be obtained from isotropic (or unpolarized) measurements.

As indicated in the article by Sprunger in this volume, angle-resolved photoemission is the most powerful method available to experimentally determine electron band structures. However, in a magnetic material, there are additional properties that are not completely resolved by measuring the band structure. Properties such as the orbital and spin moments of the electronic states and the orientations of these moments relative to crystalline axes have to be determined by other means. A thorough understanding of these properties is particularly important in today’s environment where commercial magnetic devices rely on fundamentally new materials properties exhibited in thin-film and multilayer heterostructures.

Dichroism also makes it possible to measure these quantities in an element-specific manner which is important for studies of alloys and oxides. Furthermore, one can use this effect to create images of magnetic domains,[1] as discussed by Tonner, in this volume.

Magnetic materials have a net moment that arises from the partial filling of electronic levels that result in partially filled orbitals with electrons that are not always paired in spin-up / spin-down pairs. This gives two components to the magnetic moment: the orbital component and the spin component. In 3d transition-metals, the orbital component is usually much smaller than the spin component – so-called orbital quenching. In 4d and 5d materials and 4f rare earth materials, the orbital component is significant. Many studies have been conducted to measure these two parameters since knowledge of their relative magnitudes is extremely important in understanding and predicting the underlying physics of the materials.

In this review, intended as a graduate tutorial, we will discuss the use of photon polarization to illuminate some of these properties. We will begin with a general discussion of the interaction of energetic photons with the electronic structure of atoms and solids. This will introduce the property of chirality that is exhibited in many experimental geometries. The various forms of polarized-photon spectroscopies will be enumerated. We will then discuss several commonly applied

Figure 1. Geometry in a dichroism experiment. The photon is incident with polarization vector $P$, the sample is magnetized along $M$ and the electron is emitted along $k$. These vectors can be in different planes.
methods in x-ray absorption. The final focus, however, will be on the use of polarized synchrotron light in various forms of angle-resolved photoelectron spectroscopy.

The interest in using dichroism in synchrotron studies of magnetic materials was stimulated by the discovery that photoelectron spectra of Fe 3p levels depended on whether the photons were linearly or circularly polarized, and how the polarization vector was aligned with the substrate magnetization.[2, 3] These experiments initiated a strong and ongoing series of experimental and theoretical investigations into the nature of the dichroism and into ways to predict its occurrence.

In a dichroism study, the experimental geometry must be carefully specified. In Fig. 1, we present a general diagram that illustrates the interplay of the important parameters. The incident photon of polarization $P$ can either be linearly polarized or circularly polarized. In the diagram, linearly-polarized light of either "s" or "p" orientation is shown. In the case of linearly polarized light, the relevant vector is the direction of the electric field vector $\mathbf{\varepsilon}$ while in the case of circularly-polarized light the relevant vector is the incident direction, $P$. The sample has a remanent magnetization $\mathbf{M}$ which is usually directed along a high-symmetry direction in the crystal structure. Electrons are emitted, and those detected in a photoemission experiment have a wave-vector $k$.

The origin of dichroism can be seen when one considers the photoelectron excitation cross section:

$$\frac{d\sigma}{d\Omega} = \left| \langle i | A \cdot p | f \rangle \right|^2 \delta(E_f - E_i - h\nu).$$

In this expression, the initial states are dependent on the initial wave-vector, $k_i$, which is determined by the location of that state within the Brillouin zone. Since we are considering magnetic materials, the magnetization $\mathbf{M}$ determines the nature of this initial state, $i$. An approximation that is commonly made is to re-write the differential cross-section as:

$$\frac{d\sigma}{d\Omega} = \left| \langle i | p | f \rangle \cdot A \right|^2 \delta(E_f - E_i - h\nu)$$

and now the familiar momentum matrix element, $\langle i | p | f \rangle$, where $p = -\hbar \nabla$ is clearly separated from the photon polarization. This matrix element can be calculated from a band structure calculation and it depends on the location, $k$, within the Brillouin zone where the transition takes place. This now provides the information on the coupling between $\mathbf{M}$ and $k$ in Fig. 1. The matrix elements are dependent on the direction of emission through the vector $k$ and are in general, complex. The coupling to the photon field is then given by the component $A$, above. For linearly polarized light, $A$ is aligned along the direction of the $\mathbf{\varepsilon}$ vector. Circularly polarized light carries angular momentum along $P$.

One further piece of the puzzle is provided by recognizing that the electron has spin that can be measured. In this case, knowledge of the additional spin component can provide much more information on the electronic structure. Experimentally, it is observed that ferromagnetic materials have “exchange-split” states which means that the majority spins are energetically distinct from the minority ones. Note, however, that this is quoted as an “average” splitting since it varies throughout the Brillouin zone. Bands that are pure states in high-symmetry locations become admixtures of states at other places within the zone.

II. The Different Kinds of Dichroism

Dichroism occurs when the spectrum from a magnetic material changes shape, such as a shift in binding energy and / or a change in the intensity distribution, due to a change in the sample magnetization or the photon polarization. There are two common ways to measure dichroism using synchrotron radiation: either in photoabsorption or in photoemission.
Nomenclature is important in these studies since there are many inequivalent methods that show dichroism due to different underlying physical phenomena. In photoabsorption, the photon energy is tuned through a core absorption edge, causing electrons to be excited to empty valence states near $E_F$. Absorption can be measured in many different ways such as by collecting all emitted electrons giving a total yield, measuring sample current, etc. These measurements are termed MLD and MCD (for magnetic linear dichroism or magnetic circular dichroism) when a magnetic sample is investigated with linearly- or circularly-polarized light, respectively.

In photoemission, the electron is excited to continuum energies well above $E_F$ and this results in qualitatively different information from that given by absorption. In most experiments analyzers detect the electrons in a small solid-angle, making the result specific to the electron angular distribution that is sampled. As discussed by Sprunger, changes in the photon energy and the direction of electron emission cause the measurement to sample differing regions of the Brillouin zone. This means that different initial states, with potentially different symmetries are excited. Even with imaging analyzers, discussed below, dichroism is sensitive to the angular distributions and therefore the photoemission studies are termed, in general, MLDAD or MCDAD.

Although we are interested in magnetic materials, dichroism does, in fact, occur in photoemission from non-magnetic samples and is called, for example, CDAD.[4]

**Dichroism in Photoabsorption**

In x-ray photoabsorption, the total yield from a sample is monitored as the photon energy is tuned over a core absorption edge. Usually this is done for core-levels with well-defined spin-orbit splittings such as the $2p_{1/2}$ and $2p_{3/2}$ levels of $3d$ transition-metals. The initial excitations are to empty valence states which also have well-defined spin and orbital components that are to be extracted.

When linearly-polarized light is used, the MLD signal is the difference in the spectra obtained when the magnetization, $M$, is perpendicular to the plane of polarization from that obtained when $M$ is parallel to $\epsilon$. Equivalently, one could keep $M$ fixed and rotate the polarization by 90° but this is more difficult at a synchrotron source. Note that this assumes that a remanent magnetization can be obtained at the two orientations.

As an example, shown in Fig. 2, Hillebrecht et al. found that upon reversing the vertical magnetization $M$, the total yield from Fe and Co films showed dichroism for light incident at 45° from the normal in the horizontal plane.[5] Furthermore, they demonstrated that the s-polarized light ($\epsilon$-vector in the plane of the crystal and $\parallel$ to $M$) showed no dichroism while the p-polarized light exhibited a significant asymmetry.

With the use of circularly-polarized light, MCD (or XMCD) can take advantage of sum rules that allows one to separate out spin and orbital components to the magnetic moment.[6, 7] The reason for this is that excitations from deep core levels to empty $d$-bands integrate over all of the possible excitations. The sum rules are developed from the viewpoint of an atomic transition model in which the various allowed transitions and probabilities are determined by quantum mechanics. When absorption spectra are obtained over the full energy range of the unoccupied valence bands, the resulting dichroism can be used to analyze the unoccupied states and therefore the magnetic properties. The orbital moment can be quantitatively extracted from the dichroism intensity.[6] In
principle, the extraction of the spin moment should be possible yet there is a magnetic dipole moment correction that is not necessarily small, and adds uncertainty to the process.[7] Stöhr and König have found that for 3d transition metals which have sites of lower symmetry than cubic (Oh), this correction term and the spin moment can be quantitatively separated with an additional sum rule.[8] The magnetic dipole moment is related to the anisotropy of the d charge density and they provide the appropriate correction parameters for the 5 different 3d orbitals for various alignments of the incident circular light polarization \( \mathbf{P} \) (which is given as \( \mathbf{k} \) in that paper).

In Fig. 3, the magnetic circular dichroism from Co films on Cu(001) are shown.[9] These data were obtained by measuring the photoabsorption as the photon energy is scanned over the Co 2p absorption edges. The data were taken for the photon spin \( \mathbf{P} \) aligned along the in-plane remanent magnetization \( \mathbf{M} \) for both the parallel and antiparallel conditions. This results in the characteristic dichroism signal in the Fig. 3(b). These results can be analyzed to show that the surface Co atoms have an enhanced orbital moment of 0.26 \( \mu_B \) which is twice that for bulk Co. The spin moment is found to be 1.85 \( \mu_B \) which is only 8% larger than that in the bulk.

**Dichroism in Photoemission**

Dichroism in photoemission (MLDAD, or MCDAD) has been observed in both core-level as well as valence-band excitation. The theoretical treatment of the two effects differs, however. Valence-band dichroism predictions rely on calculations of band structures and estimates of interband transitions. Theoretical treatments of core-level dichroism in photoemission, however, begin with an atomic-like description of the excitation.

The information that is obtained depends on whether one studies core, shallow core, or valence band states, since the magnitudes of the spin-orbit and the exchange (or spin) splittings vary. In the case of deep core-levels, such as the 2p’s in 3d transition metals, the ~10 eV spin-orbit splitting (i.e. the \( 2p_{1/2} \) to \( 2p_{3/2} \) splitting) is much larger than the ~0.5 eV exchange splitting; the latter results from coupling with the valence band spin polarization. On the other hand, the valence bands typically only exhibit small spin-orbit splittings (~0.2 eV) compared with their ~2 eV exchange splittings. The shallow core levels, such as the well-studied Fe 3p level, is intermediate with both energies nearly the same.

In the approach taken by Thole and van der Laan,[10] excitation of a core-level in an atom with partially-filled valence states results in a core-hole with a particular orbital and spin character which interacts with the valence spin. Since there are various ways to align the photon polarization and the emitted electron spin, there are various spectroscopies that result.[10] In combination with the potential to detect the electron spin directly, they enumerated 8 fundamental measurements that can be made.

Van der Laan goes further to outline the various independent spectra that can be measured in dichroism. In the excitation of transition-metal 2p levels to empty 3d states they find 3 independent
spectra, \( I^0 \), \( I^1 \), and \( I^2 \).[11] The \( I^0 \) spectrum is the isotropic one, \( I^1 \) is related to the “orbit spectrum,” dependent on the splitting of the \( m \) sublevels by a spin field coupled through a spin-orbit interaction. The \( I^2 \) spectrum gives an additional splitting by an electrostatic field or by magnetic alignment.

Venus warns, however that there are many inequivalent methods which differ by the orientation of \( \mathbf{M}, \mathbf{P}, \) and \( \mathbf{k} \) resulting in qualitatively different information.[12, 13] Furthermore, many synchrotron experiments are actually conducted with light that is not a pure polarization. For example, extraction of circularly polarized light off of the plane of the orbit usually leaves a linear component which can produce an (MLD) dichroism signal. For this reason, Venus favors MLDAD and suggests that other experiments (especially MCDAD as conducted by most experimentalists) be simply termed MDAD. Furthermore, many of the MCDAD experiments have an intrinsic chirality that give fundamentally different results when changing the photon handedness compared with inverting the magnetization direction.

Recently, Bansmann et al.[14] have studied this problem and despite the ambiguities pointed out by Venus, they conclude that there can be quite good agreement between theoretically and experimentally produced dichroism data, as discussed below.

Since dichroism in an angular distributions involves a comparison of intensities, it is useful to review a few relevant mechanisms that produce changes in angular distributions, including forward scattering, photoelectron diffraction, and the intrinsic band structure. Following this, we will discuss several common experimental geometries for conducting MLDAD and MCDAD.

### III. Phenomena which Modify Angular Distributions

Since our goal is to understand the origin of dichroism in photoelectron angular distributions, it is prudent to discuss various extrinsic (or post-excitation) processes which influence the intensities of these distributions. These processes can be roughly separated by the different electron kinetic energy regimes in which they are operable or dominant.

#### 500 eV and Up: Forward Focusing

When the emitted electron has energies exceeding several hundred eV, an effect called forward focussing can significantly enhance the intensity of electron emission along an atomic bonding direction.[15] The phenomena is due to the perturbation of electron trajectories by the potential of a neighboring atom, as shown in Fig. 4. Electrons are bent by the potential resulting in an excess of intensity along the direction of the bonds and a loss of intensity elsewhere.

This effect is most significant for high electron kinetic energies and for high-Z materials, which generate a larger potential for scattering. Lower energy electrons are perturbed even more by the potential and end up scattering at high angles giving rise to strong backscattering and, ultimately, photoelectron diffraction, as discussed in the next section. The energy dependence for electron scattering is shown in Fig. 5.[15]

Egelhoff has used forward focussing in XPS (x-ray photoelectron spectroscopy) as a tool to investigate epitaxy in metal-on-metal thin-film systems. By combining a study of the angle-dependence of Auger electrons and photoelectrons, it is possible to determine growth modes, study intermixing etc. The observations that are made are that, along specific emission directions, the intensity is enhanced. These emission directions are directly related to the crystalline orientation of the substrate and to the locations of the adatoms relative to those of the substrate.

![Figure 4. Forward focusing can be explained in a semiclassical manner as a perturbation of trajectories due to the potential of a neighboring atom. Ref. 15.](image-url)
Note that *multiple* forward scattering can give rise to additional contributions to this enhancement. Furthermore, features from next-nearest neighbor directions have also been observed.

As an example of this, Fig. 6 shows that when 0.5 ML of Ni are deposited on Cu, the emission of Ni $2p_{3/2}$ electrons is essentially isotropic; the drop in intensity at $\sim 80^\circ$ from the normal is an instrumental artifact. When Cu is deposited on top of the Ni, however, the Ni $2p_{3/2}$ intensity increases along a $45^\circ$ angle, showing that the Ni photoelectrons are forward scattered and the film growth is epitaxial – the Ni and Cu atoms are located in the expected places with respect to the underlying fcc Cu(001) crystal structure.

**150-500 eV: Photoelectron Diffraction**

In this regime, photoelectrons still scatter off of the potential of neighboring atoms but the lower energy increases the backscattering probability and gives rise to *photoelectron diffraction*. The wavelength of the photoelectron has dimensions of $\sim 0.5 - 1$ Å, just under the typical spacing of atoms in a crystal. This has the effect of increasing the amplitudes from multiple scattering events, and has given rise to the technique of photoelectron holography.[16] In this technique, the emission pattern from a specific atom is measured over a large solid angle. The direct photoelectron wave is the reference wave and the scattered intensities give the object wave. Interference of these waves gives rise to a pattern that can be Fourier analyzed to give the original object. The holographic reconstruction can then give a real-space pictorial representation of the geometry of the atom and its surrounding near-neighbors. In more general terms, photoelectron diffraction off of neighboring atoms can be used to extract inter-atomic spacings and local atomic geometries. Recently, bonding configurations for adsorbed C$_2$H$_2$ and C$_2$H$_4$ molecules on Si(100) have been extracted using this technique.[17] Photoelectron diffraction and its theoretical treatment has been reviewed by Fadley.[18]

Clear effects of photoelectron diffraction in MLDAD have been observed in Fe/Ag(001) and in Co/Cu(001).[19] In Fig. 7 we reproduce the third figure in Ref. [19] which shows the modulation in the intensity of the MLDAD as a function of the emission angle. The experiment was conducted with normally-incident light (pure “s” polarization) with the magnetization $\pm \textbf{M}$ in the vertical direction and electron detection in the horizontal, [11] plane. The strong oscillations are fit by a single-scattering photoelectron diffraction treatment which takes into account the various possible $(l, m)$ final-state
0-150 eV: Electronic Band Structures

Low energy photoelectrons are dominated by multiple scattering; it is essentially the same physics that is involved in photoelectron diffraction. The final state in the photoexcitation process is often represented in the simplest case to be a plane wave moving toward the detector. For simple determination of band dispersions, this approximation works rather well.

However, we are often interested in evaluating the changes in intensity that occur on reversal of the magnetization or polarization. A better representation is provided if this final state explicitly takes into account the multiple scattering — this is often termed a time-reversed LEED (low energy electron diffraction) state. In effect, this state is set up by multiple scattering from the periodic potential of the crystal. This is exactly the same kind of scattering which sets up the initial Bloch states in the crystal and should be thought of as an intrinsic property of the allowed states of that energy.

IV. Sources of Angular Distributions of Photoelectrons

In this section we will discuss some of the intrinsic sources of angular distributions and methods that are used to measure them. The focus here will be on how the photon polarization enters into producing effects on the electron angular distributions.

Angular Distributions from Unpolarized Photons

Most angle-resolved photoelectron data have been presented with minimal emphasis placed on the photon polarization. Changing the photon polarization can influence intensities, but in studies of band dispersion the emission intensity is not relevant, it is the binding energy that matters.

Here, we are interested in intensities. Although it is extremely difficult to make quantitative measures of intensity, relative intensities are straightforward to measure. There are several methods for producing intensity maps or images and we will briefly review the different methods.

The simplest method of producing an intensity map is to use unpolarized light such as that from a resonance lamp. With a commercial hemispherical photoelectron analyzer, intensity maps can be produced by scanning the analyzer over the whole hemisphere of emission angles, recording the intensity at a fixed electron kinetic energy.

In Figure 8 we show how this method can be used to measure the Fermi surface of a metal.[20] The fact that the light is unpolarized and the sample (Cu) is not magnetic means that the photoelectron emission intensities are not affected by a photon-polarization/magnetization-orientation effect. The angular distribution that is observed results from the fact that the electronic states at the Fermi level within the Brillouin zone are located only on a discrete surface of states. The intersection

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Figure 7. Angular distribution of Co 3p photoemission in 15 ML Co/Cu(001). The top panel shows the MLDAD for magnetization up and down and the lower panel shows the dichroism. The solid line is a fit using a single-scattering photoelectron diffraction theory. From Ref. 19.

of this surface of occupied states with the emitted electron’s final state (in the simplest approximation, a free-electron sphere) determines the shape of the photoemission angular distribution. In this measurement, however, the only Fermi surface contour accessible is the one shown since the photon energy of the He I resonance lamp is not tunable. Using synchrotron radiation, the region of the Brillouin zone that is sampled can be changed by tuning the photon energy, and the whole Fermi surface can be measured. However, synchrotron radiation is polarized and that means that the angular distributions are never isotropic, as discussed below.

In Figure 9, it is shown that this kind of instrument can also be used to measure x-ray photoelectron diffraction (XPD) where data from quasicrystalline Al-Pd-Mn shows pentagonal photoelectron diffraction features.[21] On the basis of a symmetry argument, solid-state physicists were confident that such a material would never exist. Nevertheless, the discovery of this structure in quench-cooled alloys has led to an upsurge in interest in such materials exhibiting only short-range order. In the lower part of the panel, XPD from the Sr and Ca core levels in Bi$_2$Sr$_2$Ca$_1$Cu$_2$O$_{8+x}$ is shown, illustrating the sensitivity to different sites in a complex crystal structure. Ref. 21, 22.

Unpolarized light has been used to observe dichroism in magnetic samples. As an example of this, in-plane magnetization reversal in Fe, Co, and Gd films on W(110) was shown to produce a clear change in intensity from normally-emitted electrons.[23] This configuration would result in no dichroism if linear s-polarized light were used with its _-vector perpendicular to M.

MUDAD (magnetic unpolarized dichroism angular distributions) relies on the ability to energy resolve the magnetic sublevels and does not rely on a spin-orbit interaction.
Polarization Dependences in Non-magnetic Systems

An alternative method to scanning over a full hemisphere of electron emission, is to measure an angular distribution over a small solid angle that corresponds to the minimal irreducible wedge of symmetry required to characterize the 2D surface. This irreducible wedge then represents the symmetry of the sample and a common practice is to symmetrize this data to produce an “angle-resolved” image of the photoelectron angular distributions.

One consideration that is missing in following the above procedure is that the matrix elements for the photoexcitation are dependent on the details of the polarization of the incident light. In Fig. 10 we present data obtained from Cu(001) which shows the photoelectron angular distributions obtained at 45, 60 and 90 eV photon energy from Cu(001) at a binding energy of 2 eV. These angular distributions were obtained from an ellipsoidal mirror analyzer[24] with the linearly-polarized light in the horizontal plane oriented at 45° from the surface normal. These data, through their left-right asymmetry, show the inherent polarization-dependent matrix elements involved in photoexcitation. These asymmetries are not observed in symmetrized data, but provide additional information on the symmetry of the initial states. For example, the state sampled at hv=90 eV in Fig. 10 must be of odd symmetry since rotation of the sample to the A || [110] condition does not turn the "x" into a "+" – rather it turns it into a "|". This additional symmetry information is extremely important in identifying the states in an unknown material.

Despite the value of the asymmetries, angular distributions are often shown from symmetrized data taken from an irreducible wedge of the sample surfaces. As an example, we could imagine taking data from Cu(001) at hv=60 eV in the irreducible wedge given with upper-left quadrant of the data taken with the polarization along [100] as given in the top-center image in Fig. 10. In that case, were we to symmetrize the image we would get the beautifully-symmetric image as given in Fig. 11(a). If, on the other hand, our experimental geometry required us to take data within the quadrant in the upper-right, our symmetrized data would look as in the image in Fig. 11(b). Clearly, these data imply drastically different conclusions regarding the location of bands within the Brillouin zone. Furthermore altogether different conclusions would be reached if the polarization were along the [110] direction as in the lower-center image of Fig. 10. In that case, symmetrization would give two totally new “angular distributions” as shown in Fig. 11(c) and (d).

This illustrates the difficulty in accepting results that are based on symmetrization of an incomplete photoemission data set. In the data in Fig 11 the details of the contours shown are not reliable representations of the initial states. There, the differences arise solely from the angular dependence of the excitation matrix elements on the photon polarization. Additional artifacts may arise as well from some of the scattering mechanisms already discussed.

Figure 10  Photoelectron angular distributions from Cu(001) at a binding energy of 2 eV for various photon energies and 2 different alignments of the incident polarization, obtained by rotating azimuthally by 45°. The left-right asymmetries are due to the linearly polarized light which is incident at a polar angle of 45° giving equal s and p components

Figure 11. Symmetrized data from the center 2 images of Fig. 10. The data in (c) and (d) have been rotated by 45° to facilitate comparison with (a) and (b).
For example, step-edge decoration of a miscut crystal may give forward focusing in one direction that is not reflected in the other, based on the crystal symmetry. We suggest that all data involving symmetrization be regarded skeptically when polarized radiation is involved in the data acquisition.

**Magnetic Linear Dichroism Angular Distributions MLDAD**

In synchrotron-based MLDAD experiments, the photon polarization is in the plane of the synchrotron orbit, which gives photons polarized horizontally. Since the polarization is usually fixed, dichroism can be observed by changing the sample magnetization, \( M \). In the case of angle-resolved detection in normal emission, reversal of a horizontal magnetization does not produce a dichroism. However, inversion of a vertical magnetization \( M \) can produce a signal. (Although it is possible to also get a difference between \( M \)-vertical and \( M \)-horizontal, theoretical treatments of this are not generally available.) The MLDAD is also predicted to have a significant energy dependence. For Fe 3\( p \), this is found to give a maximum dichroism signal around 20-30 eV (see Fig. 15, below).

As an example of the dichroism that can be seen in the valence bands, Figure 12 shows a series of photoelectron spectra measured from 6 ML Co/Cu(001) as a function of the angle \( \theta \) from the normal measured in the horizontal plane. The dichroism, plotted on the right results from reversal of the vertical in-plane magnetization. Ref. 25.

![Figure 12. Photoelectron spectra from 6 ML Co/Cu(001) as the photon incidence angle and the electron detection angle \( \theta \) are varied in the horizontal plane. The dichroism, plotted on the right results from reversal of the vertical in-plane magnetization. Ref. 25.](image1)

![Figure 13. Images of the photoelectron angular distributions obtained from 10 ML Co/Cu(001) with an ellipsoidal-mirror analyzer with linearly polarized light incident at 45° in the horizontal plane. The EDCs are shown in the top right and the dichroism in the angular distributions are shown in the lower right.](image2)
taking the difference in the two images on the left. Note that the dichroism signals are evident only in the regions where the Co intensity is observed.

This dichroism is also seen to be energy dependent as shown in Fig. 14. This is to be expected since the details of the dichroism signal are sensitive to the spin-orbit interaction. This spin-orbit interaction is dependent on the region of the electronic band structure that is sampled. Note, however that although the two images are different, they belong to the same symmetry group (C_{2v}).

Magnetic Circular Dichroism Angular Distributions MCDAD

Magnetic circular dichroism differs from linear dichroism in that a \( \Delta m_l = \pm 1 \) selection rule exists, conserving angular momentum. This changes the details of the excitations that are possible because the selection rules now have to take the orbital components, and therefore the spin-orbit coupling into account.

In synchrotron-based MCDAD experiments, the photon incidence direction, \( \mathbf{P} \), should have a component along the sample magnetization direction, \( \mathbf{M} \). The interpretation of MCDAD data rely on the specifics of the level being probed and its coupling with the valence band, and its occupation. Much of the theoretical work has been done in the context of an atomic-physics model. Therefore, specifics of the angular momentum level, \( l \), and the projections of this level, \( m \), onto the surface magnetism \( \mathbf{M} \) are crucial in the interpretation of the observed data. In effect, each \( m \) level contributes to the amplitude of an excitation and the amplitude from each depends on the orbital’s orientation relative to the light polarization. Since these are amplitudes, their relative phases enter in as well. For this reason, it is difficult to provide general selection rules that apply to a broad range of materials, and each case is best treated independently in theoretical models.

Despite this, there are several geometries that do not allow for MCDAD to occur (note that incomplete rejection of linearly-polarized light, may still allow a signal). First, a geometry, \( \mathbf{M}, \mathbf{P}, \) and \( \mathbf{k} \) completely coplanar, will result in no magnetic circular dichroism. Furthermore, a completely orthogonal geometry, \( \mathbf{P} \perp \mathbf{M}, \mathbf{M} \perp \mathbf{k}, \) and \( \mathbf{k} \perp \mathbf{P} \) will also result in a zero dichroism signal.[27]

In general, the greatest opportunity for dichroism occurs for geometries where the circularly-polarized light is incident with a component along \( \mathbf{M} \). The maximum intensity of the MCDAD signal then depends on the orientation of the electron emission direction, \( \mathbf{k} \).

Comparison of MLDAD and MCDAD

Comparison of the MLDAD and the MCDAD intensities have been made. Gerrit van der Laan has used an atomic model to explain the energy

Figure 14. The energy dependence of the MLDAD of Fig. 13. In (a) the dichroism at a binding energy of 0.4 eV is shown while (b) shows the dichroism at 0.5 eV.

Figure 15. A comparison of the predicted dichroism in MLDAD and in MCD for the Fe 3p shallow core level. (Spin-orbit ~ exchange) Note that the MCD has a minimum where the MLDAD is peaked. Furthermore the MLDAD signal, overall is larger than the MCD. Ref. 11.
dependence of the Fe 3p dichroism in both MLDAD and MCDAD. In those measurements, he predicts that MLDAD should exhibit a broad maximum peaking at 20-30 eV that MCDAD should, roughly, have a zero-crossing at the same place.[11]

This presented an experimental challenge to the community and recently Bansmann et al. have reported careful studies of this energy-dependent effect.[14]

In their experiment, they brought the synchrotron radiation to the sample at an angle of 45° from the sample normal, in the horizontal plane. Fe/W(110) films for less than 50 ML Fe had an in-plane magnetization, \( M \), in the horizontal [110] direction and the MCDAD were obtained with by inverting this magnetization. When this is done, the Fe 3p dichroism data shown in Fig. 16 are obtained as a function of the incident photon energy. These data show that the MCDAD undergoes a change in sign at \( \hbar \nu = 135 \) eV, consistent with the behavior predicted in Fig. 15, except with a significant shift in energy.

In Figure 17, the analogous data are obtained for the MLDAD condition. In this case, the in-plane magnetization had to be in the vertical direction so they used thicker films which are known to have a different direction of the remanent magnetization. When this is done, the Fe 3p dichroism data shown in Fig. 16 are obtained as a function of the incident photon energy. These data show that the MCDAD undergoes a change in sign at \( \hbar \nu = 135 \) eV, consistent with the behavior predicted in Fig. 15, except with a significant shift in energy.

In Figure 17, the analogous data are obtained for the MLDAD condition. In this case, the in-plane magnetization had to be in the vertical direction so they used thicker films which are known to have a different direction of the remanent magnetization. When this is done, the Fe 3p dichroism data shown in Fig. 16 are obtained as a function of the incident photon energy. These data show that the MCDAD undergoes a change in sign at \( \hbar \nu = 135 \) eV, consistent with the behavior predicted in Fig. 15, except with a significant shift in energy.

From these studies, it is apparent that the basic principles of MLDAD and MCDAD are established for deep core and shallow core excitations. Furthermore the application of these techniques to valence bands should be possible as well.
Spin-Resolved Detection

When the photoexcitation sensitivity to the incident polarization is combined with spin-resolved detection, we have the best opportunity to unravel all of the competing phenomena: the spin-polarization, the spin-orbit interaction and magnetic moment corrections. There are many different methods of electron spin detection including conventional Mott detectors, LEED backscattering detectors, diffuse-scattering detectors, and miniature-Mott detectors. Most rely on the asymmetry in spin-orbit scattering of the electron of the potential of the core of a heavy element. Some attempts, however, have been made to detect electron spin on the basis of the spin-polarization of empty states and the likelihood that secondary electrons are primarily one spin-polarization.

In Figure 18(a), the scattering of an electron is depicted, showing how different couplings of the spin and angular momentum give asymmetric scattering in perpendicular directions. Two recent spin detector designs are shown in Fig. 18(b) and (c). In 18(b) the NIST spin detector developed by Pierce and Celotta is shown.[28] This detector relies on diffuse scattering at ~150 eV from gold and collection of backscattered electrons in 4 quadrants. An alternate design was developed at Rice University and is essentially a mini-Mott detector.[29] Electrons are accelerated to 30 keV, strike a Au foil and the backscattered electrons are detected by 4 channeltrons. In Fig. 18(d), the LSU version of this mini-Mott detector is shown; the design is similar except for adaptation of the optics to accommodate microchannel plates rather than channeltrons. The advantage of this design is that it is compact and can be attached to a rotatable analyzer. For all spin detector designs, the counting rate is ~10^{-4} of the rate without spin analysis making it an inefficient process. Furthermore, spin detectors operated at keV accelerating potentials suffer from inadvertent arcs that make routine operation a challenge.

Spin detection is currently used to study a wide variety of problems in magnetism, including the magnetic behavior in giant magnetoresistive (GMR) films. In Fig. 19, we show angle-resolved photoemission from Cu films on Co.[30] Cu/Co multilayers are used in the magnetic bit sensor of today’s hard disk drives. The data in Fig. 19 show that quantum-well states exist in the Cu films and...
their binding energies depend on the Cu thickness. The quantum well is essentially produced by the large change in potential at the interface between Cu/Co and also by the Cu/vacuum interface.

In Fig. 20, we show Carbone’s spin-resolved data for these quantum well states.[30] Each of the peaks from Fig. 19 is found to be composed of two spin-split states with $\Delta_{\text{exchange}} = 0.2$ eV. This result is significant since it shows that Cu becomes spin polarized, and it is this effect that couples the spin information from one Co layer to another through the non-magnetic Cu spacer.

V. Summary

The use of photon polarization to control photoexcitation matrix elements presents a significant opportunity to better understand the underlying excitation mechanisms and thereby better understand the materials under investigation. We have reviewed the use of linearly- and circularly-polarized synchrotron light in the investigation of magnetic materials. We have noted that experiments can be done in both photoabsorption and photoemission and that the results that they provide are complementary. In photoabsorption there are powerful sum rules that enable one to extract quantitative values for fundamental parameters such as spin-orbit and exchange splittings. Photoemission, on the other hand provides information on these parameters as well as providing information on the spin-polarization of the electronic states and band structure within a magnetic material. With carefully-planned experimental geometries, the use of photon polarization can provide additional information that is not readily accessible otherwise.

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References