Magnetic dichroism in UV photoemission at off-normal emission: Study of the valence bands

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Magnetic dichroism of angle-resolved UV photoemission from fcc Co/Cu(001) thin films has been measured using linearly *p*-polarized light, and a coplanar geometry where the light and photoelectron wave vectors are antiparallel, and both are perpendicular to the in-plane sample magnetization. This geometry emphasizes information about state dispersion due to the crystalline symmetry. An orderly dispersion of the features in the magnetic dichroism over a wide range of *off-normal* angles of electron emission is related in detail to the bulk band structure of fcc Co. The measurements confirm the practical utility of magnetic dichroism experiments as a relatively simple complement to spin-resolved photoemission. [S0163-1829(97)04303-8]

I. INTRODUCTION

Magnetic dichroism in the angular distribution of photo-electrons (MDAD) offers the possibility of studying the electronic valence states of magnetic solids using a simple extension of widely known methods for angle-resolved photoemission. This technique involves the comparison of two photoelectron energy distributions measured for opposite senses of remanent magnetization of a magnetic sample,[1,2] and requires no special detectors or light mono-chromators (if linearly polarized or unpolarized light is used). A practical advantage of MDAD is that it is a differential spectroscopy dependent upon magnetization. It is thus ideally suited for the study of the flat, tightly spaced valence bands of the 3*d* ferromagnets near the Fermi level, where a number of photoelectric transitions may be allowed simultaneously, and the resulting peaks will overlap in the photoelectron energy distribution. The differential changes observed in MDAD may allow separation of the various component transitions and permit a more detailed study of the electronic states that support the macroscopic magnetic moment in the 3*d* ferromagnets. Such a separation of overlapping transitions was accomplished using MDAD from shallow 3*p* core states,[3] where neither the exchange splitting nor the spin-orbit interaction is large enough to be discernable in the intensity distribution, but the differential nature of MDAD reveals the internal structure of the core multiplet. The technique is in some ways similar to full spin-polarized photoemission experiments, another differential spectroscopy that gives improved state resolution through spin contrast. Spin-polarized photoemission yields, in addition, the absolute sign and magnitude of spin polarization, and can therefore provide a deeper insight into both the electronic structure and spin-dependent scattering mechanisms. However, since electron spin polarization measurements are time consuming and require a rather specialized apparatus, MDAD has a role as complementary technique, which permits a rapid survey of the electronic structure with some degree of spin sensitivity. Detailed spin-polarized photoemission experiments could then pursue selected experimental conditions of particular interest.

Little systematic work investigating the valence bands of ferromagnets using MDAD has been reported thus far. A number of groups have reported the observation of MDAD using ultraviolet light,[4] and have shown that the dichroism is of order 1% to 5% without correction for the effects of the secondary electron background. There has been little analysis of these results, with the emphasis being instead on quantifying the size of the dichroism and confirming its symmetry properties. A few very recent studies have demonstrated the utility of MDAD in *normal* photoelectron emission. One such work investigates nickel films on Cu(001), as a function of the photon energy.[5] It uses circularly polarized light and a highly symmetrical geometry where the magnetization, photoelectron, and light wave vectors are all parallel to the surface normal. The experimental separation of the small spin-orbit splitting of valence bands of different double-group symmetry is impressive, as is the agreement with quantitative one-step photoemission calculations of the MDAD. A second work[6] presents an analysis of a single MDAD spectrum for normal emission from the in-plane magnetic system Co/Cu(001). It uses unpolarized light and off-normal illumination, and demonstrates the ability to detect a point of band hybridization. One-step photoemission calculations are again in good agreement, but predict an absolute spin polarization that cannot be understood, and that MDAD does not address.

The present paper investigates the practical usefulness of MDAD for valence-band mapping in emission directions of

low symmetry, that is, for *off-normal* photoelectron emission. Despite expectations, the one existing study describing the angular dependence of MDAD is discouraging: significant magnetic dichroism from Co/Cu(001) using unpolarized He *I* radiation is observed only for normal emission,[7] and there is no indication that the technique will be useful for following band dispersion away from a high symmetry line. This is particularly surprising, since previous results for the angular dependence of MDAD from core levels is encouraging: even for emission from deeper ($2p$) core levels of the $3d$ ferromagnets, the influence of the crystalline symmetry is observable[8]; in the case of the shallower $3p$ levels, the effect of crystallinity on the final state (alternatively viewed, diffraction effects) is strong.[9] It is not clear why MDAD of the valence bands should be an effective technique for normal emission, but not for off-normal emission. One-step photoemission calculations of MDAD for emission along directions of low symmetry are not yet available, but a recent qualitative analysis[10] suggests that this should not be the case. MDAD is expected whenever there is initial state hybridization of band states by the spin-orbit interaction. It is also expected in any dipole-allowed transition due to spin-orbit coupling in the final state (often termed spin-dependent surface transmission[11]). These initial and final state effects should have distinct signatures. The present experiments therefore revisit the Co/Cu(001) system, and use the systematic dependence of MDAD on photon and photoelectron wave vector, crystalline orientation, and light polarization to choose an experimental geometry that emphasizes magnetic dichroism due to band effects. Significant MDAD in off-normal emission for Co/Cu(001) is observed, and is shown to have a systematic dispersion related in detail to band-structure calculations.

II. EXPERIMENTAL PROCEDURE

Face-centered cubic Co films with thicknesses of 5–6 ML were grown epitaxially by atomic deposition onto a Cu(001) single-crystal substrate at room temperature.[12] The growth and thickness were monitored using medium-energy electron diffraction oscillations and Auger electron spectroscopy, and the film quality was assessed by low-energy electron diffraction. The films were remanently magnetized using an electromagnet with a soft iron core, and the magnetic state was characterized by measuring a hysteresis loop using the magneto-optical Kerr effect (MOKE). It was found that, for this substrate crystal, the in-plane magnetization was rather independent of magnetization axis, and that a robust remanent magnetization along [100] was stable for long times. This behavior has been previously reported,[12] although it is more common for the remanent axis to be along [110] for these films.[13] The photoelectron distributions were measured in an adjoining chamber, where the magnetic field from the magnetizing solenoid was carefully screened. The magnetic dichroism measurements were therefore performed as a series of spectral measurements (each occupying a few minutes) followed by a magnetization reversal. Upon every reversal, MOKE was used to confirm both that the sample had held its original remanent magnetization and that the remanent state had been reversed for the subsequent measurements.
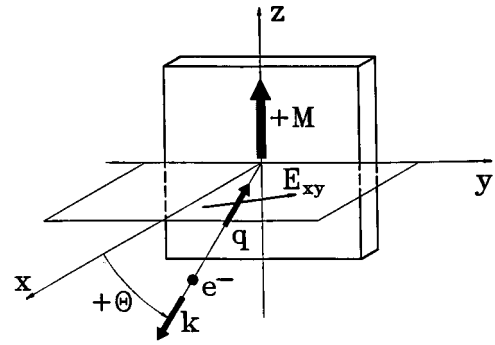

FIG. 1. The Co fcc film is remanently magnetized along the z axis, and has crystalline mirror planes in both the xz and xy planes. Linearly polarized light is incident with wave vector **q** in the xy plane at an angle θ to the surface normal. The polarization vector $\mathbf{E}_{xy}$ is also in the xy plane. Photoelectrons with wave vector $\mathbf{k} = -\mathbf{q}$ are collected in an angle- and energy-resolving spectrometer and the energy distribution is measured for both senses of **M**.

Figure 1 illustrates the geometry of the angle-resolved photoemission measurements. Linearly polarized light from the BESSY synchrotron source was incident at angle θ to the surface normal, with both the light wave vector **q** and the polarization vector lying in the synchrotron plane and normal to the magnetization **M**. The cylindrical-mirror electron spectrometer[14] had a hole in the outer cylinder that allowed the light to pass through the spectrometer and electron lens, and thus allow analysis of photoelectrons with wave vector **k** antiparallel to **q**. This geometry has a number of advantages. First, models of magnetic dichroism based on emission from oriented atoms predict no effect in this geometry when linearly polarized light is used.[2] Since models based on crystalline symmetry predict nonzero MDAD even in this geometry where **k**, **q**, and **M** are coplanar,[10] it is expected to emphasize information about the electron bands and their dispersion due to crystallinity. Second, the experimental geometry is predicted to have specific symmetry properties as the angle θ is varied. Since the xz plane is a mirror plane R_v of the crystalline structure, the dichroism upon magnetization reversal obeys[10]

$$D^M(\mathbf{k}=-\mathbf{q}, r=1, \alpha=\pi) = -D^M(\mathbf{k}'=-\mathbf{q}', r=1, \alpha=-\pi),$$

(1)

where r and α denote the ellipticity and orientation of the polarization ellipse ($r=1$, $\alpha=\pm\pi$ for linear polarization in the xy plane), and $\mathbf{k}\rightarrow\mathbf{k}'$, $\mathbf{q}\rightarrow\mathbf{q}'$, $\theta\rightarrow-\theta$ under reflection in R_v. This reversal of the dichroism upon reversal of the angle θ allows an independent check of the correctness of measurements of small asymmetries that could be sensitive to a number of experimental artifacts. Third, variations in θ may be accomplished by sample rotation about a single axis, thereby minimizing the difficulties of comparing data taken with different arrangements of the spectrometer and monochromator optics. Finally, linearly polarized light is used to enhance the surface sensitivity.[10] There are two disadvantages to this experimental geometry. First, it is not possible to study normal emission, since according to Eq. (1) the magnetic dichroism will be zero. However, the experiments mentioned previously[5,6] have shown that these experiments present no problem under different conditions. Second, the light passing
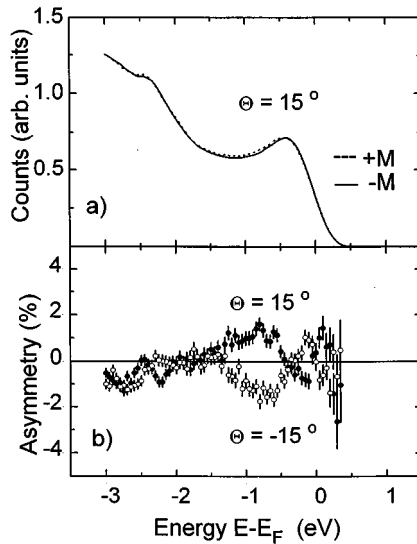
FIG. 2. (a) Photoelectron intensity distributions are shown for $\theta = 15°$ and a photon energy of 16 eV, for both senses of remanent magnetization. The magnetic dichroism is defined as the difference of the two curves divided by their sum, and is shown by the solid symbols in part (b). The magnetic dichroism measured in this way for $\theta = -15°$ is shown as the open symbols in part (b).

through the spectrometer produces more secondary electrons than would normally be present, and results in a large secondary electron background in the intensity spectra at larger binding energies. This effectively limits the dichroism measurements to within a few eV of the Fermi energy. This, however, is precisely the region of interest.

Sample magnetic dichroism measurements are presented in Fig. 2. A photon energy of 16 eV was used, and $\theta = 15°$. The intensity distributions in part (a) show a peak close to the Fermi energy due to the Co d bands, and a further smaller peak at about -2.5 eV that arises from overlapping Co and Cu bands. The strong secondary electron background previously mentioned is also evident. The two distributions for the two senses of remanent magnetization show small differences that are quantified in part (b) of the figure, where their normalized difference is plotted as an asymmetry in solid symbols. Since the observed intensity asymmetry upon magnetization reversal is only a few percent, it is prudent to verify that it is not the result of experimental artifacts. To this end, the measured asymmetry for data measured for $\theta = -15°$ is also plotted using open symbols. According to Eq. (1), the magnetic dichroism should reverse sign as θ is reversed. The data show that this is indeed the case for binding energies less than about 2.5 eV, and that reproducible MDAD features exist at ≈ -0.1 and ≈ -0.75 eV. Near -1.7 eV there is no MDAD within experimental error, but below ≈ 2.5 eV there is a small asymmetry that does not reverse upon reversal of θ. The latter is likely an artifact due to the secondary electrons produced in the spectrometer.

These results confirm that MDAD using UV light is experimentally practical, even though the observed asymmetries are rather small. Furthermore, it illustrates how MDAD due to band-structure effects is present in the geometry where $\mathbf{k} = -\mathbf{q}$ is coplanar with $\mathbf{M}$, even though an atomic model appropriate to emission from core levels predicts a null result.
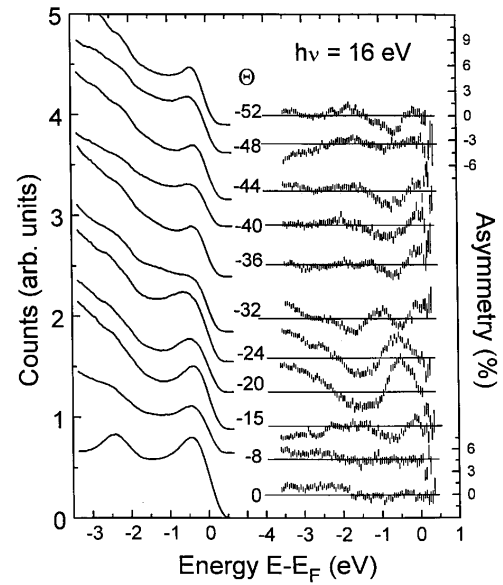

FIG. 3. A series of photoelectron energy distributions (left-hand axis) and magnetic dichroism curves (right-hand axis) are presented for angle-resolved photoemission with linearly polarized light with a photon energy of 16 eV. The angle θ shown in Fig. 1 is varied from curve to curve. The curves are displaced for clarity; zero lines for the dichroism are shown explicitly, and those for the intensity distributions are indicated by the data above about 0.5 eV.

III. VALENCE-BAND DISPERSION

The angular variation of the MDAD was measured for a wide range of angles θ, with a constant photon energy of 16 eV. The data for negative values of the angle are displayed in Fig. 3. On the left is the average intensity distribution, and on the right is the corresponding intensity asymmetry. The intensity distributions are unremarkable—it would be very difficult to discern band dispersion within the broad feature near the Fermi level using a traditional analysis. One could only say that the shape of the peak changes somewhat with θ, leading to a suspicion that more than one transition is involved. However, the MDAD measurements on the right of the figure give a clear quantification of these subtle shape changes, and, since the asymmetry is due to the reversal of the sample magnetization, imply that they have a magnetic origin. There is also significant magnetic dichroism in the region from -1 to -2 eV, implying the presence of transitions that are not discernable as peaks in the intensity distributions. The features in the MDAD show definite dispersion as would be expected from band dispersion as $\mathbf{k}_\parallel$ (electron wave vector parallel to the surface) is varied.

The magnetic dichroism data are replotted in Fig. 4 as a function of the binding energy and parallel wave vector $k_\parallel$. $k_\parallel$ is normalized to $\mathbf{k}_x$, the distance from Γ to X in the bulk Brillouin zone. An individual spectrum taken at constant emission angle traces a curved line on this plot. The contour plot in Fig. 4 is made by interpolating between the twelve data sets in Fig. 3 and another five sets for positive values of θ. Solid contour lines denote positive asymmetry, while dotted lines denote negative asymmetry. The asymmetry interval between contours is 0.5%, and the zero contour is not plotted to aid in the clarity of presentation. The overall structure of the plot is in agreement with Eq. (1). In the range of
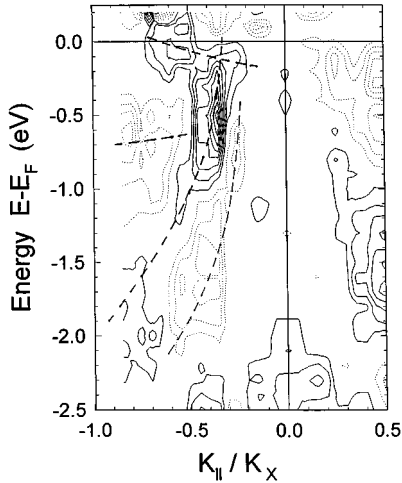
FIG. 4. Contours of constant magnetic dichroism interpolated from the data in Fig. 3 are plotted as a function of binding energy and electron wave vector parallel to the surface. Positive contours are shown as solid lines; negative contours are shown as dotted lines; the zero contour is not plotted. The interval between contours is 0.5%. The dashed lines are guides to the eye, which delineate the dispersion of features of maximum magnetic dichroism.

$k_\parallel$ where a comparison can be made, reversal of $k_\parallel$ changes the sign of the asymmetry without affecting the main shape of the contours. There is some disparity in the absolute asymmetry for $\pm k_\parallel$, as can already be seen from Fig. 2. However, noting the size of the statistical error bars in that figure, a difference of a single contour line is close to the size of the error bars. There are also a few features in the plot that do not show the expected reversal of asymmetry. These are either above the Fermi level, where the statistical uncertainty in the asymmetry is large, or in a semicircular region bounded by $E \lesssim -2$ eV, $|k_\parallel/k_x| \lesssim 0.5$, where the secondary electron background can produce a false asymmetry as previously discussed with reference to Fig. 2.

Concentrating on the contours for negative $k_\parallel$, there is unambiguous dispersion of the major regions of positive and negative dichroism with $k_\parallel$. Approximate lines following the maxima and minima of the magnetic dichroism contours have been sketched in as dashed lines to aid in the following discussion. A positive maximum disperses rapidly from $k_\parallel/k_x = -1$, $E \approx -2$ eV until it crosses the Fermi level at $k_\parallel/k_x = -0.25$. It is accompanied by a negative maximum that disperses along a roughly parallel line, displaced to a lower energy. The displacement is in the range of 0.9–1.2 eV, depending on which angle is chosen in the asymmetry data for its evaluation. This pair of features is suggestive of minority and majority spin bands separated by an exchange splitting. Since both the photoelectron wave vector and the light polarization vector lie within the xy mirror plane, these bands must be of even reflection symmetry if dipole transitions are to be allowed. In order to see if this interpretation is reasonable, it would be best to compare the dispersion of these features to one-step photoemission calculations of MDAD, which include both the exchange interaction, the spin-orbit interaction, and all other relativistic effects in a consistent, nonperturbative manner. Such calculations exist for some systems,[5,6,15] but not for the low symmetry directions probed in these experiments. A more qualitative analy-
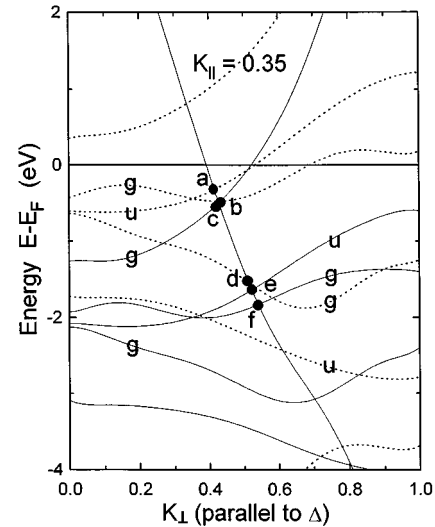

FIG. 5. The bulk band structure for fcc Co is presented for a line parallel to Δ, which lies in the xy mirror plane, and has $k_\parallel/k_x = 0.35$. Solid lines indicate majority bands and dotted lines indicate minority bands. Many bands are labeled by the letters g, for the even-group representation, or u for the odd-group representation. The thick solid line is a minority-majority spin pair of bands with negligible exchange splitting and even spatial symmetry, which has been translated down in energy by 16 eV. The intersections a through f mark possible bulk photoelectric transitions.

sis may be performed using a two-step model of photoemission and the widely available band structure calculations, which neglect relativistic effects.[10] Figure 5 presents a spin-polarized, nonrelativistic, calculation of the bulk band structure of fcc Co, along the low symmetry line parallel to Δ, in the xy mirror plane, with $|k_\parallel/k_x| = 0.35$. A nonrelativistic version of the Koringa-Kohn-Rostoker method described in Ref. 16 was used. The solid curves on this plot denote majority (spin down) bands and the dotted curves denote minority (spin up) bands. Many of the occupied bands have been labeled as either "g" (for Γ_g, the even representation) or as "u" (for Γ_u, the odd representation). The thick solid line shows a pair of final-state bands of even symmetry, opposite spin, and negligible exchange splitting, which have been shifted down by 16 eV. The intersections marked by the letters a through f indicate possible bulk photoelectric transitions. Indeed, the intersections labeled b and f in Fig. 5 represent transitions from a pair of exchange-split bands of even symmetry, which occur at initial-state energies in good agreement with the energies at which the two magnetic dichroism extrema in Fig. 4 cross through $k_\parallel/k_x = -0.35$. The magnetic dichroism observed for these bands is likely due to the presence of spin-orbit coupling in the final state, which in the context of an analysis based upon bulk states, is spin-dependent transmission at the surface.

The other two extrema in magnetic dichroism may also be interpreted as dispersing initial-state bands. A second positive maximum starts above the Fermi level at large $k_\parallel$, crosses it at $k_\parallel/k_x = -0.6$, and continues to smaller $k_\parallel$ with rather flat dispersion until it intersects the rapidly dispersing minority band discussed above. The energy position at $k_\parallel/k_x = -0.35$ suggests that this is the transition labeled a in Fig. 5, and that both positive maximum contours represent

minority bands. Transition *a* is from a band of odd symmetry, and would not be observed in the absence of spin-orbit coupling. However, MDAD exists only because of the presence of spin-orbit coupling, so its effects should be considered. In a mirror plane normal to the magnetization, spin-orbit coupling hybridizes only states of opposite spin and opposite parity. The observation of transition *a* implies that a transition to a majority band of even symmetry must also be allowed with very similar coordinates in $k_\parallel$ and energy. Then the two states will be hybridized by spin-orbit coupling, so that both bands have a sizeable admixture of even symmetry character and both transitions should be observed. The magnetic dichroism observed at such hybridization points is an initial-state effect most analogous to MDAD observed from emission from core states. The signature of such a hybridization point should be a plus/minus feature in the magnetic dichroism. Reference to Fig. 5 shows that transition *c* has precisely the required attributes. In fact, the convergence of transitions *a*, *b*, and *c* at $k_\parallel/k_x = -0.35$ creates overlapping contours of magnetic dichroism that are difficult to interpret. However, moving to more negative $k_\parallel/k_x$ in Fig. 4, it is evident that the final dispersing negative maximum of magnetic dichroism at approximately -0.7 eV, which merges with the others at $k_\parallel/k_x = -0.35$, in fact represents the majority band of even symmetry which leads to transition *c*. As predicted, the magnetic dichroism is negative, so that the two hybridized bands form a plus/minus asymmetry pair with small energy separation.

The contour plot does not give clear evidence of bands leading to the possible transitions *d* and *e*. These two bands of opposite spin and parity would also be expected to hybridize via spin-orbit coupling and transitions from both would be allowed. The fact that they are not observed in the magnetic dichroism may be due to one of two reasons. These bands approach each other at a much sharper angle in Fig. 5 than do the hybridizing bands at smaller binding energy. It may be that they will hybridize strongly and be observable over a shorter range of $k_\parallel$, and within this range are masked by the large dichroism due to the strongly dispersing majority band, which yields transition *f*. Alternatively, it may be that they are more difficult to detect at larger binding energy because of the increased secondary electron background.

The measured contours of magnetic dichroism may also be compared to a previous result at normal emission. The present measurement geometry of $\mathbf{k} = -\mathbf{q}$ gives no dichroism for normal emission along Δ, but it is reasonable to extrapolate the observed minority and majority features in the dichroism so that they intersect $k_\parallel = 0$ at approximately $E = -0.3$ and -0.6 eV, respectively. Spin-resolved photo-emission experiments for normal emission from similar fcc Co/Cu(001) films,[17] using a photon energy of 16 eV, reveal minority and majority peaks at these energies, and so are at least consistent with the present results.

IV. CONCLUSIONS

MDAD using linearly polarized light proves to be a practical and useful method to study the valence bands of $3d$ ferromagnets in directions of low emission symmetry. Use of an experimental geometry where $\mathbf{k} = -\mathbf{q}$ appears to emphasize the magnetic dichroism arising from band states and discriminates against magnetic dichroism, which has its origin in atomic models. Because of its differential nature, the magnetic dichroism reveals structure within rather broad photoemission intensity peaks and allows overlapping transitions to be detected. Transitions are also revealed within a large signal of secondary electrons. The features in the magnetic dichroism show an orderly dispersion with $k_\parallel$ that has been related directly to calculations of the bulk band structure. The origin of these features can be understood qualitatively with reference to widely available band-structure calculations for $3d$ metals, which do not include spin-orbit coupling. Because of the complicated nature of photoemission, this approximate form of analysis cannot be expected to hold true in all cases—still, the present results verify the predictions of the qualitative model and encourage the development of more sophisticated calculations of MDAD in geometries of low symmetry. Magnetic dichroism arising from both initial-state effects (band hybridization by spin-orbit coupling) and final-state effects (spin-dependent surface transmission) can be identified, and are of the same magnitude for emission in this mirror plane. These results hold in principle for MDAD in off-normal emission using unpolarized light as well, but there are important issues of detail to be addressed. Unpolarized light contains both linear polarizations, each with half the relative intensity. This should allow more dipole transitions, but the asymmetry from each will be, on average, half as large. It is not yet clear whether or not this will produce so many overlapping, weak magnetic dichroism features, that not even MDAD is useful for the separation of the individual transitions.

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