

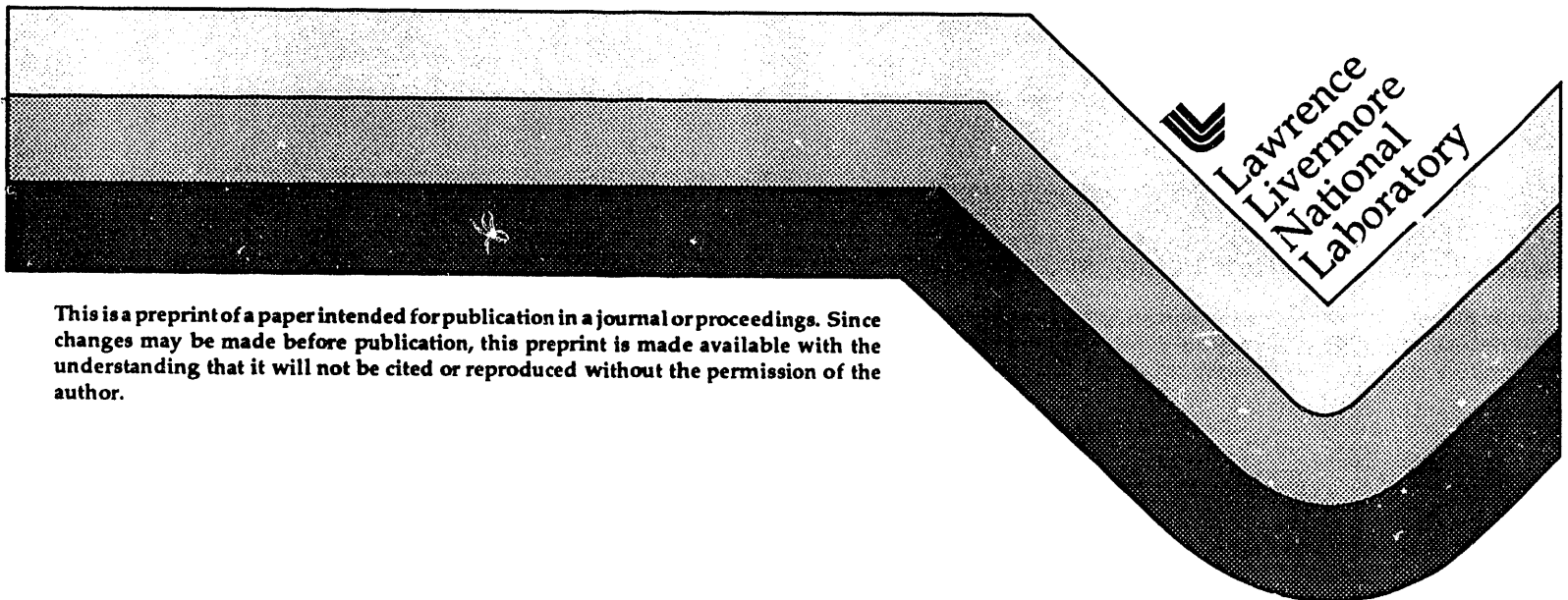
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## Magnetic Circular Dichroism in X-Ray Absorption and Core-Level Photoemission

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### ABSTRACT

Here is reported observation of magnetic circular dichroism in both x-ray absorption and core-level photoemission of ultra thin magnetic films using circularly polarized x-rays. Iron films (1–4 ML) grown on a Cu(001) substrate at 150 K and magnetized perpendicular to the surface show dramatic changes in the  $L_{2,3}$  branching ratio for different x-ray polarizations. For linearly-polarized x-rays perpendicular to the magnetic axis of the sample the branching ratio was 0.75. For films  $\geq 2$  ML, this ratio varied from 0.64 to 0.85 for photon spin parallel and anti-parallel, respectively, to the magnetic axis. This effect was observed either by changing the x-ray helicity for a fixed magnetic axis, or by reversing the magnetic axis for a fixed x-ray helicity. Our observation can be analyzed within a simple one-electron picture, if the raw branching ratios are normalized so that the linear value becomes statistical. Furthermore, warming the films to  $\sim 300$  K eliminated this effect, indicating a loss of magnetization in the film over a temperature range of  $\sim 30$  K. Finally, reversing the relative orientation of the photon spin and the magnetic axis from parallel to anti-parallel allowed measurement of the exchange splitting of the Fe 2p and 3p core levels which were found to be 0.3 eV and 0.2 eV, respectively. These results are consistent with earlier studies, but the use of off-plane circularly-polarized x-rays from a bending magnet monochromator offers  $\sim 2$  orders of magnitude greater intensity than typical spin-polarization measurements. Finally, we have performed preliminary x-ray absorption studies of  $UFe_2$ , demonstrating the feasibility of MCD measurements in 5f as well as 3d materials.

One of the basic thrusts of the investigation of nanoscale magnetic structures, whether it be ultrathin monolayer films, multilayers, or clusters, is the establishment of structure-property relationships. To be more specific, the determination of the interconnections between geometric, electronic, and magnetic structure is the fundamentally important but intermediate goal, with an eye towards the ultimate manipulation and application of this understanding. Ordinarily, efforts to measure nanoscale magnetic properties in conjunction with atomistic geometric and electronic structures runs headlong into the same problem: The magnetic perturbation tends to be a small component of the overall effect. In contrast to this, here we report giant circular dichroism in the near-edge core-level x-ray absorption of a near-monolayer metal film. An example of our data is shown in Fig. 1. The essential effect is the relative amplification of the  $2p^{1/2}$  peak where the magnetization and helicity are parallel, regardless of whether the magnetization is into or out of the surface plane. (Because the magnetization and x-ray incidence are normal to the surface, we

call it perpendicular dichroism.) This is a direct measurement of the spin polarization and the density of the unoccupied states near  $E_F$  in a ferromagnetic system.

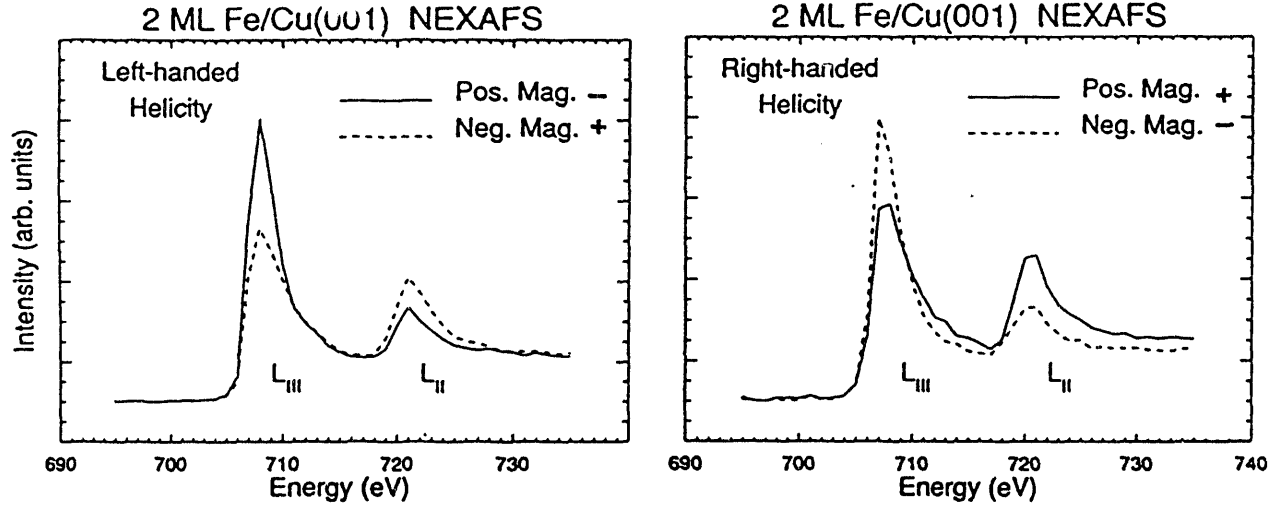


Figure 1. The near-edge x-ray absorption fine structure (NEXAFS) dichroism of 2ML of Fe/Cu(001). These are plots of absorption versus photon energy. The upper panel shows the effect of reversing the magnetization while maintaining the left-handed helicity of x-rays. Similarly, for the lower panel and right-handed helicity x-rays. Samples are perpendicularly magnetized either into (pos. mag.) or out of (neg. mag.) the surface. The symbol +(-) means that helicity and magnetization are parallel (anti-parallel). The  $-2p_{3/2}$  peak is at the  $L_{III}$  edge and the  $2p_{1/2}$  peak is at the  $L_{II}$  edge. The spectra were normalized to each other by equating the pre-edge intensity, at energies below approximately 700 eV.

The temperature dependence of the perpendicular magnetization can also be followed with this technique. Figure 2 illustrates the behavior for 4 ML of Fe/Cu(001). Here, we are using branching ratio (BR) as a measure of the dichroism. BR is defined in Eq. (1).  $I$  is the integrated intensity of the white line peak at each edge jump.

$$BR = I(2p_{3/2}) / [I(2p_{3/2}) + I(2p_{1/2})] \quad (1)$$

We have developed a simple, one-electron picture to analyze our results, which can be summarized into the closed form analytic expression shown in Eq. (2).

$$BR = \frac{3 + 2(1 - \alpha)\beta + 2(1 - \beta)\alpha}{6} = \frac{2}{3} - \frac{(P_{lv})(P_e)}{6} \quad (2)$$

Previously, we expressed this equation in terms of  $\alpha$  and  $\beta$ .  $\alpha$  is the spin down alignment of the unoccupied 3d states and  $\beta$  is the degree of right circular polarization. We have now derived a more compact form, using the classical definitions of polarization. See Table I for details.

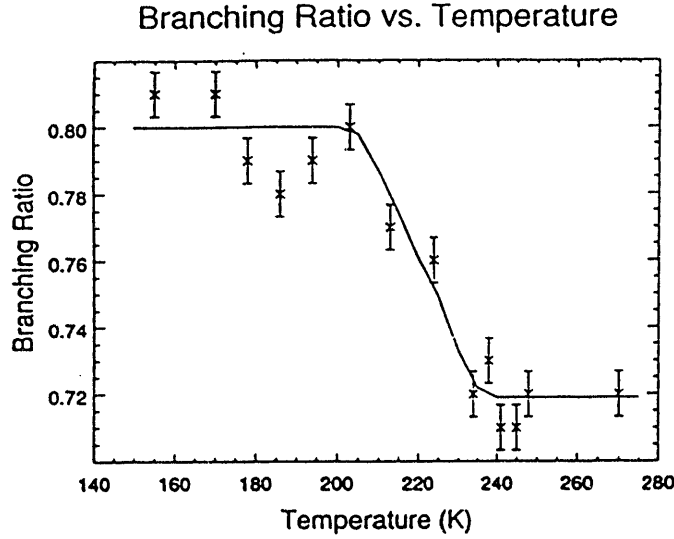


Figure 2. The temperature dependence of the magnetization of 4 ML Fe/Cu(001) using NEXAFS magnetic circular dichroism as the probe. Because of the imperfect placement of the thermocouple on the sample plate, the sample temperature may be slightly higher than that shown. Error bars of approximately  $\pm 1\%$  are included for comparison but the true time error estimate is probably higher (about 4%). A branching ratio near 0.72 corresponds to an unmagnetized sample.

	Empty Valence States		
	Pure Down	Pure Up	Random
$\alpha = \frac{N_{\text{UNOCC}}^{\downarrow}}{N_{\text{UNOCC}}^{\downarrow} + N_{\text{UNOCC}}^{\uparrow}}$	1.0	0.0	0.5
$P_e = \frac{N_{\text{UNOCC}}^{\downarrow} - N_{\text{UNOCC}}^{\uparrow}}{N_{\text{UNOCC}}^{\downarrow} + N_{\text{UNOCC}}^{\uparrow}} = 2\alpha - 1$	1.0	-1.0	0.0
	Photon Helicity		
	Pure Right Circular	Pure Left Circular	Linear
$\beta = \frac{I_R}{I_R + I_L}$	1.0	0.0	0.5
$P_{\text{hv}} = \frac{I_R - I_L}{I_R + I_L} = 2\beta - 1$	1.0	-1.0	0.0

Table I:  $N_{\text{UNOCC}}$  is the number of empty d valence states. The symbols  $\uparrow$  and  $\downarrow$  denote spin up and down, respectively.  $\alpha$  is the spin alignment of the d valence states.  $P_e$  is the polarization of the d valence states.  $\beta$  is the degree of right circular helicity of the x-rays.  $P_{\text{hv}}$  is the circular polarization of the x-rays.  $I_R(I_L)$  is the intensity of the right (left) circularly-polarized component of the x-ray beam.

To use a one-electron picture, it is necessary to "normalize-out" the non-statistical many-body effects. To do this we multiply all BR values by  $2/3/BR_{EXP}(lin)$ . This allows us to concentrate upon the effects of helicity variation. The results of our analysis, assuming  $2\mu_B/Fe$  atom ( $\alpha = 25\%$  and  $75\%$ ) is shown in Table II.

Table II.

P versus M	BR <sub>EXP</sub>	BR' <sub>EXP</sub>	BR <sub>TH</sub>
+	0.645	0.585	0.60
-	0.83	0.75	0.73
lin	0.74	0.67	0.67

P = polarization

M = magnetization direction

P vs M: relative directions of helicity and magnetization

+

= parallel

-

= anti-parallel

BR = branching ratio

BR<sub>EXP</sub> = raw experimental results

BR'<sub>EXP</sub> = experimental results, normalized to the linear statistical prediction

BR<sub>TH</sub> = atomic theory prediction,  $2\mu_B/Fe$  atom,  $\alpha = 25\%$ ,  $75\%$  and  $\beta = 10\%$ ,  $90\%$  pol.

We have also used MCD with core-level photoemission to measure the exchange splitting of the  $2p_{3/2}$  and  $2p_{1/2}$  peaks of 4ML of Fe/Cu(001)<sup>2</sup> as well as the Fe3p peak of 2 ML of Fe/Cu(001). The spectra are shown in Figures 3 and 4. The exchange splitting of  $2p_{3/2}$  peak is  $0.22 \pm 0.10$  eV. The apparent change in the spin-orbit splitting between the two peaks is  $0.33 \pm 0.14$  eV. The exchange splitting of the Fe3p is approximately 0.2 eV. There are less than those observed in the bulk (see Reference 2 and references therein). All measurements were made in remanence and with perpendicular magnetization.

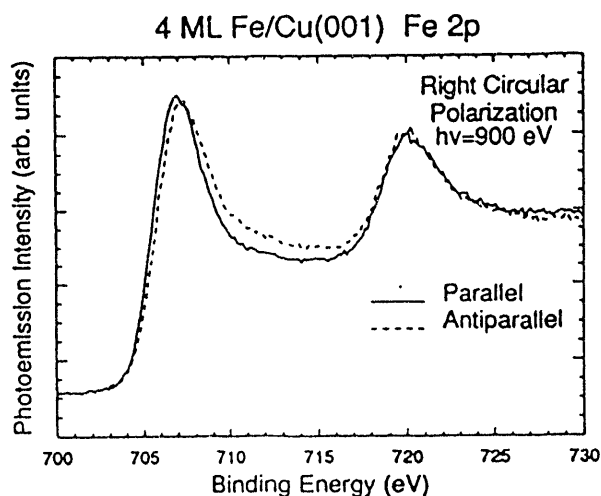
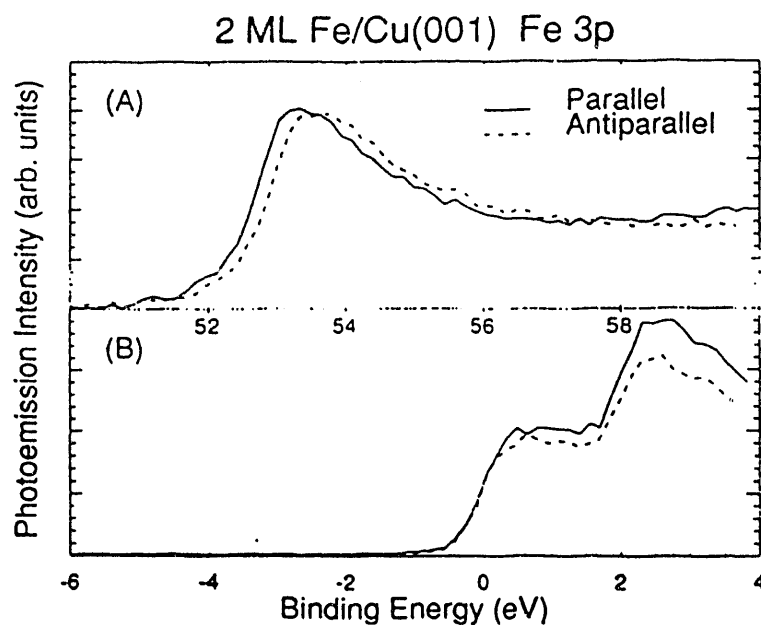
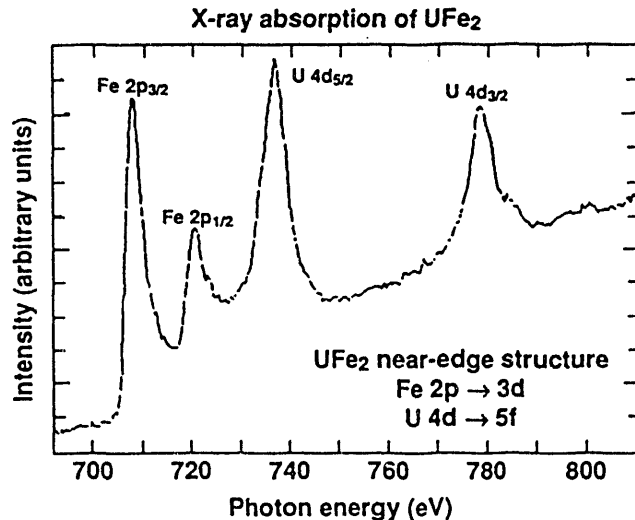


Figure 3 Fe 2p spectra taken with  $h\nu = 900$  eV and positive photon helicity. The solid lines are for parallel orientation of photon spin and sample magnetization and the broken lines for an anti-parallel orientation. The two orientations were achieved by fixing the photon helicity and reversing the magnetization of the sample. The spectra are as collected and show the differences in binding energy for the  $2p_{3/2}$  and  $2p_{1/2}$  levels for the two helicity and magnetization orientations.



**Figure 4** Magnetic circular dichroism in photoemission of the fcc-Fe 3p core level. The bottom panel shows the alignment of the Fermi edge, which allows for a direct comparison of the 3p binding energies in the top panel. The sample was 2 ML Fe/Cu(001), with perpendicular magnetization.



**Figure 5** The near-edge x-ray absorption fine structure (NEXAFS) spectrum of UFe<sub>2</sub>, taken with linearly-polarized x-rays, is shown here. Note the strong transitions for both the Fe2p and U4d states.

Recently we began a series of experiments using near-edge-x-ray-absorption-fine structure (NEXAFS) looking for magnetic-circular-dichroism (MCD) in the magnetic actinide materials UFe<sub>2</sub> and U-S. These and our previous experiments were performed at SSRL using the University of California/National Laboratories PRT beamline 8-2<sup>3</sup>. This SGM beamline

permits us to access the  $2p \rightarrow 3d$  transition of Fe ( $B^F = 707, 720$  eV) and the  $4d \rightarrow 5f$  transition of U ( $B^F = 736, 778$  eV). Absorption experiments with circularly-polarized x-rays should allow direct, elementally-specific interrogation of the magnetic and related electronic structure of these materials, in analogy with the results from magnetic x-ray scattering<sup>4</sup>. Our initial experiments utilized NEXAFS<sup>(1)</sup>, but in the future they will also be expanded to include core-level photoemission<sup>2</sup> as well.

We expect that the orders-of-magnitude improvement in brightness at the ALS will open the door for an unprecedented opportunity for both basic science using 5f elements, as well as improving the overall understanding of the physical and chemical properties of uranium compound materials.

We have performed magnetic circular dichroism (MCD) experiments upon Fe/Cu(001), using both core-level photoemission and near-edge absorption fine structure. A simple, one electron picture worked well to explain normalized branching ratio measurements. We plan to continue and expand this work, including pursuing the measurement of MCD effects in 5f systems.

### Acknowledgments

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