Magnetic x-ray dichroism in 2*p* absorption spectra of Fe/Cu(001)

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Large magnetic circular dichroism using circularly polarized synchrotron radiation has been observed at the $L_{2,3}$ absorption edges of thin (1–12 monolayers) fcc Fe films grown on Cu(001). Dramatic changes in the 2*p* branching ratio are observed when the orientation of photon helicity and sample magnetization are varied from parallel to antiparallel. The temperature and film thickness dependence of the perpendicular anisotropy in these films could be monitored by variations in the 2*p* branching ratio. Finally, our results are described in a simple theoretical framework that allows a determination between the predicted low- and high-spin phases for fcc Fe. Our data suggest a high-spin phase with a moment of 2.0–2.5 μ_B/atom. Interestingly, thicker films with remanant magnetic moments in the film plane present smaller branching ratio variations consistent with either a reduced moment or with domain closure for these films. The surface sensitivity and elemental specificity of this technique make it particularly attractive for the study of surface and thin film magnetism.

The recent availability of highly circularly polarized photons in the soft x-ray regime provides a unique opportunity for the study of magnetic materials. Pioneering efforts have used the circularly polarized x-rays for x-ray absorption[1,2] and core-level photoemission.[3,4] Each of these studies measures the relative change in cross section as the incident x-ray polarization is varied. These changes are referred to as x-ray magnetic circular dichroism.

In the x-ray absorption measurements, varying the x-ray polarization from right to left circular gives rise to relative intensity changes in the L_2 and L_3 absorption edges for 3*d* transition metals and in the M_4 and M_5 edges for rare earths. The results are qualitatively explained by an exchange-split valence-band model.[5] Simple single-electron atomic calculations which include crystal-field interactions can be used to extract an approximate local magnetic moment from the absorption spectra.[2,6] Furthermore, in addition to offering elemental specificity, when collected in an electron partial yield mode, the absorption spectra also offer surface sensitivity and can be used to estimate local moments for magnetic surfaces and thin films.

In this paper, we show results of x-ray absorption measurements for the Fe/Cu(001) system. This system has been studied by a number of techniques including spin-polarized electron spectroscopies,[7] inverse photoemission,[8] surface extended x-ray-absorption fine structure (SEXAFS),[9] low-energy electron diffraction,[10] x-ray photoelectron diffraction,[11] and surface magneto-optic Kerr effect (SMOKE).[12]

All films in this study are grown at ~150 K where Fe forms a relatively poorly ordered metastable fcc film.[11] At low coverage and low temperature, the perpendicular anisotropy of these films exceeds the magnetic dipole energy giving rise to a magnetic easy axis normal to the surface. This perpendicular anisotropy has a complex temperature and coverage dependence which can be followed by observing the switching transition between perpendicular and planar remanent magnetization.[7]

All measurements were performed at the Stanford Synchrotron Radiation Laboratory (SSRL) using the spherical grating monochromator that is part of the facilities of the University of California–National Laboratories Participating Research Team.[13] It is well known that radiation just above and below the electron orbital plane in the storage ring is highly circularly polarized.[14] The capability of this monochromator to produce usable intensities of highly circularly polarized x rays was first demonstrated by Wu *et al.*[15] The degree of x-ray polarization for the results presented here is estimated to be ~90%.[16]

The Fe was evaporated from a W basket located ~50 cm from the sample. Typical evaporation rates, as monitored with a quartz crystal microbalance, were ~1 ML/min. at pressures $\leqslant 10^{-9}$ Torr. Fe coverages were further calibrated using the relative intensities of the Fe and Cu 3*p* core levels which were cross calibrated with previous Auger electron spectra from this system.[17]

The Fe film was magnetized by applying current pulses to a coil whose axis could be oriented either perpendicular to or along the sample surface. The maximum applied field was ~3 kOe and all measurements were made in remanence.

X-ray magnetic circular dichroism in the 2*p* absorption spectra of 3*d* transition metals is a consequence of the dipole transition selection rules. It can easily be shown that excitation with left-circularly polarized x rays permits only transitions with $\Delta m_j = -1$, and right-circularly polarized x rays only those with $\Delta m_j = +1$. Thus, in the case where large initial-state spin-orbit splitting exists the absorption spectra with right- and left-circularly polarized x rays will be different. More precisely, the spectra will depend upon the relative orientation between the x-ray helicity vector and the sample magnetization.

Examples of Fe 2*p* absorption spectra for 2 ML Fe/

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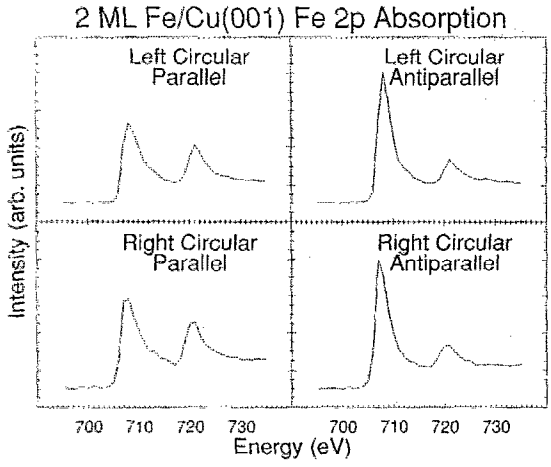

FIG. 1. X-ray absorption spectra for 2 ML Fe/Cu(001) at 150 K. The left panels show results for parallel helicity and sample magnetization, and the right panels show the same for the antiparallel alignment. The four extremes of alignment, i.e., left- and right-circularly polarized x rays for a magnetization both into and out of the sample, are shown. In each case the Fe $2p$ branching ratio changes from 0.83 for parallel photon helicity and sample magnetization to 0.64 for antiparallel photon helicity and sample magnetization.

Cu(001) at 150 K are shown in Fig. 1. One immediately notices the dramatic difference in relative intensities of the $2p_{3/2}$ and $2p_{1/2}$ manifold when the orientation of the photon helicity and sample magnetization are changed from parallel (left panels) to antiparallel (right panels). For these films at this temperature the magnetic easy axis is along the surface normal and these spectra were acquired with normal photon incidence.

Table I summarizes the experimental x-ray absorption data for 2 ML Fe films at 150 K and compares them with a simple one-electron atomic theory which parallels the pioneering efforts of Erskine and Stern.[5] The results are expressed as a branching ratio which is defined as the percentage of the $2p$ absorption intensity in the $2p_{3/2}$ manifold. Within this model, this value is $\frac{2}{3}$ for linear polarized x rays or nonmagnetized samples. The experiment gives a value of 0.74 for linearly polarized x rays which can be explained by incorporation of spin-orbit interaction in the valence band. This, of course, produces an orbital component to the total magnetic moment. It has, however, been demonstrated that for Fe, Co, and Ni the orbital moment is $\sim 10\%$ or less of the spin moment.[18] Our approach is to remove these effects by normalizing the experimental data to the linear results. The implicit assumption is that the "nonstatistical" effects are not dependent on the x-ray polarization.[2] It is important to note that recent calculations have demonstrated that x-ray absorption dichroism directly measures the shell-specific orbital moment[19] and thus provides the opportunity for assessing both the orbital and spin contributions to the total magnetic moment. Currently, extraction of the spin moment is not as direct as for the orbital moment. The results herein represent a simplistic approach predicated on the relatively small magnitude of the orbital moment and the applicability of atomic models to these films. The two theoretical branching ratio columns in Table I are for moments of 4 and 2 μ_B/Fe and x-ray polarization of 100%. The agreement between experiment and theory for 2 μ_B/Fe is quite remarkable. A slightly more sophisticated analysis[20] which includes adjustable x-ray polarization and electron spin populations in the unoccupied d states yields the following expression for the magnetic moment (MM) of Fe:

$$\mathrm{MM}(\mu_B) = -8\left(\frac{3\,\mathrm{BR}' - 2}{P}\right),$$

where BR $'$ is the normalized experimental branching ratio and P is the x-ray polarization. From our data with $P = \pm 0.90$ we get ~ 2.2 μ_B/Fe. The accuracy of this determination is probably no better than ± 0.5 μ_B/atom. Nevertheless, in the case of fcc Fe films, it allows us to distinguish between the predicted low- and high-spin phases with moments of ~ 1 μ_B/atom and 2.5 μ_B/atom.[21]

We now turn to the temperature dependence of the perpendicular anisotropy of the Fe films. We use the $2p$ branching ratio taken with circularly polarized x rays to follow the loss of perpendicular magnetization as the sample is warmed. If, for example, at 150 K the photon helicity and sample magnetization are antiparallel and the $2p$ branching ratio is ~ 0.83, then as the sample is warmed above ~ 200 K the branching ratio decreases until at ~ 230 K it reaches a final value of ~ 0.72. This indicates that the magnetic easy axis has switched from normal to the surface to parallel to the surface. This transition is reversible provided the sample is not warmed above ~ 350 K, and these results are consistent with earlier electron spin-polarization[7] and SMOKE[12] measurements.

Finally, we turn to the coverage dependence of the perpendicular anisotropy. In Fig. 2 we present results for

TABLE I. 2 ML Fe film. Comparison of experimental and theoretical branching ratios. The normalized experimental values are obtained from the experimental values by multiplying by (0.67/0.74). The theory values are for Fe moments of 4 μ_B and 2 μ_B and 100% circular polarization.

			Branching ratio			
Polarization	Magnetization	Orientation	Exp.	Norm. Exp.	4 μ_B Theory	2 μ_B Theory
Left	Out	Parallel	0.64	0.58	0.50	0.58
Left	In	Antiparallel	0.83	0.75	0.83	0.75
Right	In	Parallel	0.65	0.59	0.50	0.58
Right	Out	Antiparallel	0.83	0.75	0.83	0.75
Linear			0.74	0.67	0.67	0.67

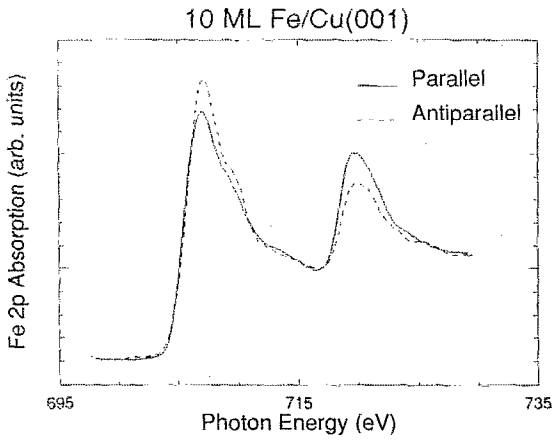

FIG. 2. X-ray absorption spectra for 10 ML Fe/Cu(001) at 150 K. Sample magnetization was along the surface, and the spectra were acquired for 10° grazing photon incidence. The solid (dashed) curves are for helicity and magnetization approximately parallel (antiparallel).

10 ML Fe/Cu(001) at 150 K. This film is fcc, but its magnetic easy axis is now along the surface. Table II shows that the branching ratio for 10° glancing incidence (left side) varies from 0.66 to 0.74 for photon helicity and sample magnetization approximately parallel and antiparallel respectively. On the right side of Table II we show the branching ratio as a function of incident photon angle to demonstrate the dependence of the branching ratio on the alignment of photon helicity and sample magnetization. Again, using the simple theory of Refs. 2 and 20, and compensating for the misalignment of photon helicity and sample magnetization, we get a reduced value for the Fe magnetic moment of $\sim 1.4\ \mu_B$/atom. This result is consistent with a recent study of a 15 Å fcc Fe film on Cu(001) (Ref. 7) which postulated a reduced moment for these films. However, it is also possible that the apparent reduction in the Fe moment is due to domain closure in these films. For the films with perpendicular magnetization macroscopic single domains should exist for sufficiently high perpendicular anisotropy.[22]

We have presented results that exploit the availability of circularly polarized soft x rays in the study of magnetic materials. The x-ray absorption measurements probe the spin-split density of states just above the Fermi energy and contain information about the magnetic moment of the sample. Simple one-electron theories are sufficient to produce semiquantitative agreement with experiment. This method combines elemental specificity and surface sensitivity and is therefore particularly suited to the study of surface and thin-film magnetism. It is complementary to existing spin-polarized photoemission techniques and its use should become increasingly prevalent in the study of $3d$ and $4f$ magnetism.

TABLE II. 10 ML fcc Fe film. Fe $2p$ branching ratio for a thick fcc film with magnetic easy axis along the surface. The left side shows the variation for 10° glancing incidence as the orientation of the photon helicity and sample magnetization is changed from approximately parallel to antiparallel and compared to normal incidence (perpendicular). On the right side we show the branching ratio as a function of photon incidence angle. The normalized experimental branching ratio is obtained as in Table I.

Orientation	Exp.	Norm. Exp.	θ	Exp.	Norm. Exp.
Parallel	0.66	0.63	10°	0.66	0.63
Antiparallel	0.74	0.70	55°	0.68	0.65
Perpendicular	0.70	0.67	90°	0.70	0.67

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