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X-ray magnetic linear dichroism in absorption at the L edge of metallic Co, Fe, Cr, and V

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It is demonstrated that x-ray magnetic linear dichroism (XMLD) in absorption spectroscopy is a viable technique for element-specific magnetic characterization of *metallic* thin films and multilayers. XMLD is measured at the Fe, Co, Cr, and V $L_{2,3}$ edges, and varies from 1–11% of the edge jump in the absorption coefficient, with a magnitude that scales with the square of the magnetic moment. The XMLD spectra show satisfactory agreement with first principles calculations of this effect. [S0163-1829(98)50832-6]

Because of its element specific nature, synchrotron based x-ray magnetic dichroism[1–3] has become an important tool for the study of magnetic moments in alloys and multilayers. The most common dichroism variants (magnetic circular dichroism in photoemission (MCD),[4] x-ray magnetic circular dichroism in x-ray absorption (XMCD),[1,3] and magnetic linear dichroism in the angular distribution of electrons (MLDAD),[5] are sensitive to the average magnetization $\langle M\rangle$ of a particular species and are useful for the study of ferromagnetic or ferrimagnetic systems. A second class of dichroism techniques are sensitive to $\langle M^2\rangle$ [specifically, magnetic linear dichroism in photoemission (MLD),[6] the transverse magnetooptic Kerr effect (T-MOKE) (Ref. 7) and its x-ray absorption analog,[8] and x-ray magnetic linear dichroism in x-ray absorption (XMLD) (Refs. 2 and 9)]. These provide greater opportunity for the study of antiferromagnetic (AF) systems.

Here we focus on XMLD, which measures changes in the x-ray absorption coefficient. As such, it does not require an electron spectrometer, and is experimentally less demanding than photoemission techniques such as MCD, MLDAD, and MLD. Furthermore, XMCD and XMLD spectra are now well understood in terms of sum rules[10–13] which permit their decomposition into components arising from $\langle S_z\rangle$, $\langle L_z\rangle$, etc. As compared with XMCD, XMLD has the additional advantage that it uses linearly polarized radiation, since the most commonly used synchrotron radiation sources are linearly polarized, and the extraction of circularly polarized radiation from such sources usually incurs a significant loss of intensity.[14]

However, until now there have been only two reports using XMLD[2,9] as compared with dozens using XMCD, partly because most work has focused on ferromagnetic systems. In a ferromagnetic transition metal[15] the XMCD fraction of the absorption coefficient $[\approx(n_+-n_-)/(n_++n_-)]$ is larger than the XMLD fraction $\{\approx[(n_+-n_-)/(n_++n_-)]^2\}$ [here $n_{+(-)}$ is the hole occupation number of the spin up (down) bands]. For example, in bulk Fe the peak XMCD at the $2p$ edge is $\approx 150\%$ (normalized to the edge jump), while we show here that the peak XMLD is only $\approx 8\%$.

Another subtle difference between XMCD and XMLD is elucidated by Ref. 12. There dichroism spectra are decomposed into components arising from $\langle S_z\rangle$ (labeled w^{011}), $\langle L_z\rangle$ (w^{101}), and so forth. At the L_2 edge of Fe, both $2p_{1/2}$ multiplets contribute positively to the $\langle S_z\rangle$ component of the XMCD. Comparatively, these multiplets have equal and opposite contributions to the $\langle S_z\rangle$ component of the XMLD. Therefore to measure $\langle S_z\rangle$ with XMCD, one needs only to distinguish the L_3 and L_2 edges (separated by 13 eV in Fe). The same measurement by XMLD is more difficult, since it requires resolution of the individual multiplet states (separated by ≈ 1 eV).

To make matters worse, each multiplet has an intrinsic broadening that is inversely proportional to the core-hole lifetime. In the solid state the atomic multiplets are further broadened into bands. Thus, if the core-hole lifetime is short or the hybridization strong, the $\langle S_z\rangle$ component of the XMLD spectrum is strongly suppressed. This is exactly the situation in transition metal (TM) systems, hence XMLD may not be measurable in a metal.[16] Indeed, the two extant studies reporting nonzero TM XMLD have focused on metal oxides,[2,9,17,18] where the multiplet peaks are narrow and well separated.[20]

Here we present the first XMLD spectra taken at the L edge of Co, Fe, Cr, and V *metallic* thin films and multilayers.[19] These are shown to be in satisfactory agreement with first principles calculations of this effect. We verify the XMLD equivalent of the ''transferability approximation'' [22] in XMCD, wherein the magnitude of the XMLD (when normalized to the edge jump) is proportional

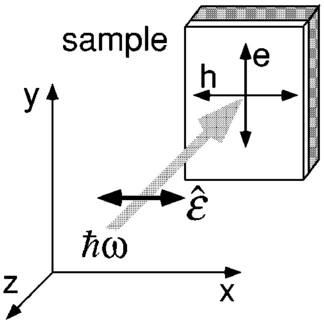

FIG. 1. The x rays strike the sample surface at normal incidence. The easy (e) and hard (h) axes are perpendicular and parallel, respectively, to the in-plane component of the applied magnetic field (Ref. 28). X-ray absorption spectra are taken with $\langle M \rangle$ parallel and perpendicular to the polarization axis $\hat{\varepsilon}$.

to $\langle M^2 \rangle$. Also presented is an approach for the measurement of XMLD spectra that simplifies experimental requirements by taking advantage of magnetocrystalline anisotropy in epitaxial thin films.

This paper focuses on four samples prepared by sputter deposition (as described elsewhere[23]) with structures:

A, 25 Å Fe/200 Å Fe/50 Å FeCo/50 Å Co @ 250° C;
B, 25 Å Fe/200 Å Fe @ 300 °C;
C, 25 Å Cr/15 Å Cr/[1.5 Å Cr/7.5 Å Fe]$_{40}$@100 °C;
D, 25 Å Cr/15 Å Cr/[1.5 Å V/7.5 Å Fe]$_{40}$@100 °C;

MgO(110) substrates were used. The first 25 Å layer of either Fe or Cr was grown at 600 °C to enhance pseudo-epitaxial growth and each film was capped with 20-Å Al to prevent oxidation. Specular x-ray diffraction showed that the films were epitaxial, with a bcc (112) orientation (see Refs. 23 and 24), except for the topmost Co layer of sample A, which had an hcp (100) texture.[25] All samples showed a strong uniaxial magnetic anisotropy. Easy and hard magnetic axes were determined by magneto-optical Kerr effect (MOKE) magnetometry (see, e.g., Refs. 23 and 24) and hard axis saturation fields were all $\lesssim 1000$ Oe. Each sample showed negligible remanence along its hard axis, indicating a 90° change in magnetization direction between saturation and zero applied field.[26]

Since XMLD is maximized when the magnetization is switched by 90° between parallel and perpendicular to the photon polarization, $\hat{\varepsilon}$, it usually requires two electromagnets. Here we eliminate one electromagnet by exploiting the magneto-crystalline anisotropy[27] in our epitaxial films, as shown in Fig. 1. Samples are mounted with the magnetic hard axis, (h), parallel to the polarization axis, $\hat{\varepsilon}$, which is also parallel to the x component of the applied magnetic field,[28] $\vec{H}_x$ ($\mathrm{h} \| \hat{\varepsilon} \| \vec{H}_x \| \hat{x}$). The sample easy axis, (e), is aligned along $\hat{y}$. The synchrotron radiation strikes the sample with normal incidence. This nullifies any signal arising from circular dichroism or T-MOKE.[29] Since the sample is fixed relative to the incident beam and the magnetization is rotated (XMLD of the first kind), these measurements have no contribution from the crystal field.[9,21]

The measurements were made at the Synchrotron Radiation Center in Madison, Wisconsin. Photoabsorption experiments were made using $>90\%$ linearly polarized light from a bending magnet source with a total electron yield technique. Data were taken in applied magnetic fields sufficient to saturate the magnetization along the hard axis ($|\hat{M} \cdot \hat{\varepsilon}| = 1$, $\vec{H}_x \leqslant 1000$ Oe), and in zero applied field with the magnetization relaxed to the easy axis ($|\hat{M} \cdot \hat{\varepsilon}| = 0$).

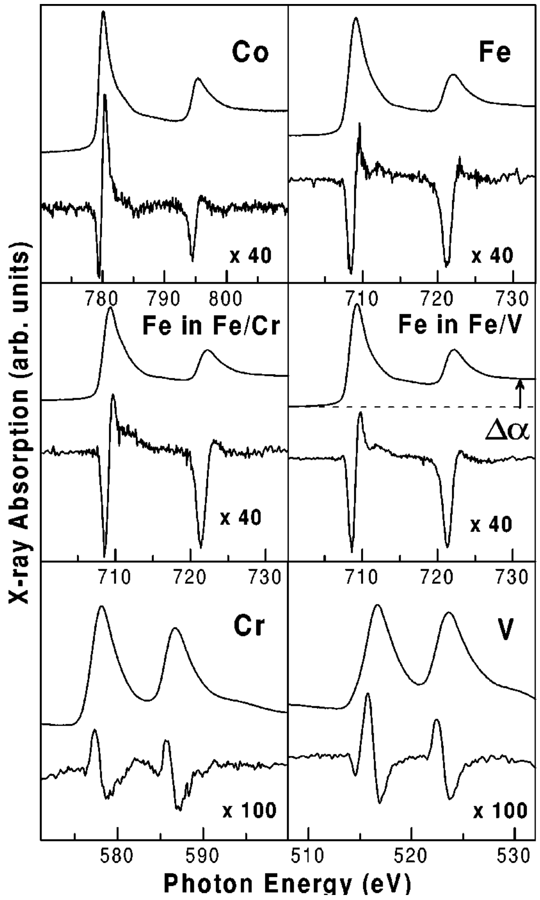

FIG. 2. X-ray absorption spectra for Co (sample A), Fe (B, C, and D), Cr (C), and V (D) at the $2p$ absorption edges. In each case, the lower curve shows the XMLD spectrum obtained from the absorption spectra, scaled as indicated.

To neutralize circular dichroism effects due to sample misalignment, or any other potential asymmetry in the apparatus, measurements were taken in the sequence of $H^+, 0^+, H^-, 0^-, \ldots,$ applied field. Here $H^{\pm}$ refer to $\vec{H}_x$ applied along the positive or negative x axis, while $0^{\pm}$ indicates the remanent state following saturation in the $\pm$ directions. Magnetization directions were switched at each photon energy. The absorption spectra H^+ and H^- (or 0^+ and 0^-) were averaged to give the absorption spectra $\alpha^{\|}(\hbar\omega)$ [or $\alpha^{\perp}(\hbar\omega)$].[30]

Figure 2 displays the absorption curves at the Co, Fe, Cr, and V $2p$ edges. The linear dichroism, defined as $(\alpha^{\perp} - \alpha^{\|})$, is clearly measurable in all cases. To obtain a quantitative measure of the XMLD, we define

$$\mathrm{XMLD}_{\max} = \max \left| \frac{\alpha^{\perp} - \alpha^{\|}}{\Delta\alpha} \right|, \tag{1}$$

where $\Delta\alpha$ is the step height far above the L edges (see example in Fig. 2) as described in Refs. 31 and 22 (dichroism signal scaled to a per-atom basis). This particular (but arbitrary) choice was made since an integral of the XMLD over

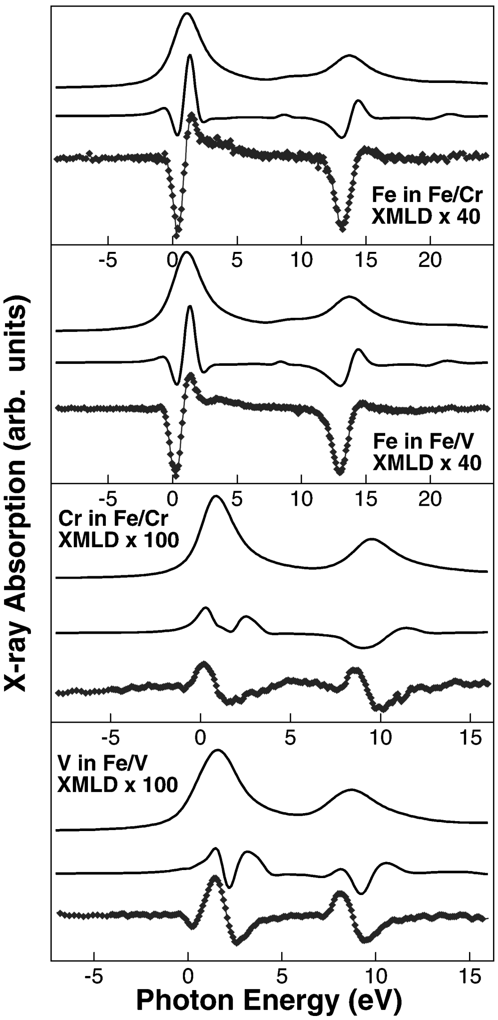

FIG. 3. Theoretical x-ray absorption and XMLD spectra (solid lines) from the structures discussed in the text. For comparison, the respective experimental XMLD spectra are plotted on the same scale (markers).

any edge would identically cancel out the contribution from the spin or orbital moments.[12] Furthermore, we found experimentally that because of the difficulty of getting a perfectly flat baseline with in-field measurements, XMLD integrals could not be reliably distinguished from zero to within experimental error.

Theoretical XMLD spectra (Fig. 3) were calculated within the itinerant one-electron model from the spin-polarized relativistic band structures of bcc Fe_5Cr_1 and Fe_5V_1 (112) multilayers.[20,25] The band structures were calculated self-consistently using the relativistic linear muffin-tin orbital method based on the first principles local spin-density functional theory (LSDA) [see Refs. 20 and 21 and references therein]. The orbital polarization as implemented in Ref. 32 which corrects the errors in the orbital magnetic moments given by the standard LSDA calculations, was included. Ideal bcc (112) superlattices were assumed and the experimental bcc Fe lattice constant was used for the in-plane lattice constants.

For the Fe_5/V_1 multilayer, the Fe-V interlayer spacing was expanded by 8.3% to take into account the larger V atomic volume. The calculated total magnetic moments are $2.27\mu_B$ (Fe), $-0.70\mu_B$ (Cr) for Fe_5/Cr_1, and $2.20\mu_B$ (Fe), $-0.87\mu_B$ (V) for Fe_5/V_1. The calculated magnetic anisotropy energies show that both multilayers have a uniaxial in-plane anisotropy with the easy axis along [110] and the hard axis along [111] in agreement with experiment. The theoretical absorption and XMLD spectra plotted in Fig. 3 have been Lorentzian broadened to take the finite core-hole lifetime into account. The broadening used here is slightly smaller than that used in previous calculations,[21] which reflects the high photon energy resolution of the experimental measurements.

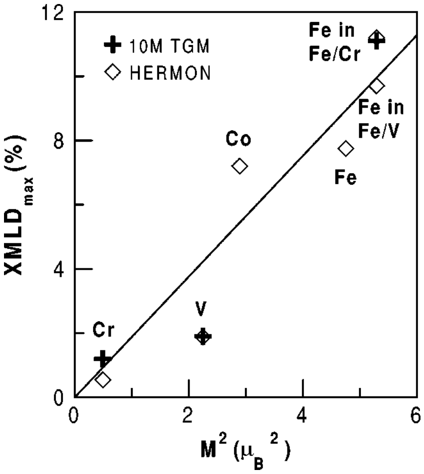

FIG. 4. $XMLD_{max}$ [Eq. (1)] is compared with $\langle M^2\rangle$ for the elements studied here. The two kinds of symbols represent two sets of measurements taken on different beamlines with different photon energy resolutions. The line fit indicates $XMLD_{max} = 1.88\cdot M^2(\mu_B^2)$ with correlation coefficient $R^2 = 0.96$. While Cr and V are not normally magnetic at room temperature, they have previously been shown to acquire the stated magnetic moments in multilayers with Fe (Refs. 23 and 24).

The overall magnitude and shape of the theoretical Fe and V spectra (Fig. 3) agree reasonably well with the measured ones. Further calculations showed that the shapes of the Cr and V XMLD spectra are sensitive to the Fe-Cr or Fe-V interlayer spacing. Thus, the differences in the shapes of the experimental and theoretical Cr or V XMLD spectra may reflect the structural differences between the experimental and theoretical multilayers.

The sum rules for XMLD provide no information regarding the spin or orbital moment.[12] To quantify $\langle M^2\rangle$ measurements, we propose an approach analogous to the determination of $\langle M\rangle$ from XMCD in Refs. 23, 24, and 33. First, the absorption curve from a sample is compared with that of a standard, and they are put on the same scale. Then the sample XMLD magnitude is compared with that of the standard, resulting in a measure of $\langle M^2\rangle$. For this process to succeed it is important that the XMLD spectrum does not change in shape but only in magnitude between the sample and standard. In Fig. 2 it is seen that the three Fe spectra studied here have the same shape, which bodes well for this approach.

Another useful analysis of XMCD spectra uses the ‘‘transferability approximation’’ [22] whereby it is possible to compare magnetic moments in *different* elements by comparing suitably scaled XMCD spectra. This method is especially helpful in systems where the number of valence band holes is not known. In Fig. 4 we test the analogous approximation for XMLD. The experimental $\mathrm{XMLD}_{\max}$ for each element is plotted versus $\langle M^2 \rangle$, as determined by XMCD. $\mathrm{XMLD}_{\max}$ is shown to be highly correlated to $\langle M^2 \rangle$, in agreement with expectation.

For the first time, XMLD is measured in metallic transition metals. Signal levels up to 11% of the absorption step height are obtained. A comparison with theory shows favorable agreement. Since XMLD measures $\langle M^2 \rangle$ it is especially favorable for antiferromagnetically coupled systems. As noted above, XMLD spectra have much more fine structure than analogous XMCD spectra. Thus XMLD spectra provide a more difficult test for theory, and the combination of high quality experiment and theory promises to reveal more information regarding the spin-polarized band structure in the investigated metals.

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[14] Elliptical or helical undulators, on the other hand, are very effective sources of circularly polarized light.

[15] Note that this statement is not necessarily true for rare earth materials, see, e.g., J. B. Goedkoop, B. T. Thole, G. van der Laan, G. A. Sawatzky, F. M. F. de Groot, and J. C. Fuggle, Phys. Rev. B **37**, 2086 (1988).

[16] By comparison, XMLD effects are much larger in the gas phase, where the orbital moment is not quenched. See, e.g., A. von dem Borne *et al.*, Phys. Rev. Lett. **78**, 4019 (1997).

[17] One previous study of bulk Ni found negligible XMLD [J. Vogel and M. Sacchi, Phys. Rev. B **53**, 3409 (1995)]. This was probably due to the small Ni magnetic moment and the relatively high noise levels in that experiment.

[18] Several studies have reported XMLD in rare earth systems, see, e.g., J. Vogel and M. Sacchi, J. Magn. Magn. Mater. **150**, 293 (1995); Surf. Sci. **365**, 831 (1996).

[19] XMLD has two realizations.[20,21] XMLD of the ''first kind'' (employed here) measures the change in absorption coefficient while the magnetization direction is alternated between parallel and perpendicular to the photon polarization. This is a purely magnetic effect, and is directly proportional to $\langle M^2 \rangle$. XMLD of the ''second kind'' holds the magnetization fixed while the photon polarization is alternated. XMLD of the second kind contains contributions from nonmagnetic sources, such as of the quadrupole moment of the electron charge distribution.

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[25] The crystal structure of the cobalt film was not sufficiently well characterized to undertake a calculation of the XMLD to compare with this sample. Since preliminary calculations (Refs. 20 and 21) have already shown that the XMLD effect can depend on the crystal structure, we don't expect the present experiment to agree with previous results.

[26] Although remanence in these films may not be a single domain state, the predominant orientation of the magnetization is along the easy axis (apart from the rather small volume in the domain boundaries). Since XMLD is sensitive only to $\langle M^2 \rangle$, the multidomain nature of the film is unimportant.

[27] We point out that while the uniaxial magnetic anisotropy was employed to rotate the magnetization, it is not a necessary requirement for XMLD. We have recently observed the XMLD effect in fourfold symmetric Fe(100) and Ni(100) samples when magnetized along their easy axes [M. M. Schwickert *et al.* (to be published)]. Furthermore, we emphasize that XMLD is readily observed in thick films (>1000 Å) hence is a ''bulk'' effect, and is not predicated on the symmetry breaking of a surface or interface.

[28] The apparatus was originally designed for XMCD, hence the magnetic field was actually applied at 45° with respect to the surface normal when the sample is aligned perpendicular to the incident x-ray beam. However, due to large demagnetization fields in these samples, the magnetization never departed from the sample plane by more than 4°.

[29] Recall that T-MOKE is the change in the reflection coefficient of p-polarized x rays depending on the magnetization direction (Ref. 8).

[30] Due to a difference in detection efficiency for in-field and remanent measurements, $\alpha^{\parallel}(\hbar\omega)$ were multiplied by a constant factor to put them on the same scale as $\alpha^{\perp}(\hbar\omega)$. Additionally, a quadratic polynomial curve was sometimes subtracted from the dichroism spectrum, whenever the difference between $\alpha^{\parallel}$ and $\alpha^{\perp}$ spectra deviated in the region before the L_3 or far beyond the L_2 edges. Such corrections were usually small, and the resulting spectra were highly reproducible, even on different beamlines.

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