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Soft-x-ray magnetic circular dichroism at the $L_{2,3}$ edges of nickel

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Magnetic circular dichroism (MCD) has been observed at the $L_{2,3}$ absorption edges of ferromagnetic nickel by use of circular-polarized soft-x-ray synchrotron radiation. The MCD intensity ratio between the L_2 and the L_3 edges is found to differ appreciably from that predicted by a simple exchange-split-valence-band model. Fine MCD features, imperceptible in the absorption spectra, are also observed and a tentative interpretation is given. This work, demonstrating the feasibility of MCD measurements in the soft-x-ray region, provides a new approach to study $3d$ and $4f$ ferromagnetic systems with their respective dipole-permitted $2p \rightarrow 3d$ and $3d \rightarrow 4f$ transitions.

Synchrotron radiation from the bending magnets of electron storage rings has the desirable property that the radiation emitted just above or just below the plane of the ring has a high degree of circular polarization. In the ultraviolet region, this property has been exploited in spin-polarized photoemission measurements on gases and solids,[1] and in circular dichroism and magnetic circular dichroism (MCD) studies of both inorganic and biological molecules.[2] Starting with a paper by Erskine and Stern,[3] there has been increasing agitation to perform dichroism-type experiments with synchrotron radiation in the soft-x-ray region. The attractive feature of this region is that it admits access to the strong dipole-permitted (core $2p$) $\rightarrow$ (valence $3d$) excitations in transition-metal ferromagnets, and (core $3d$) $\rightarrow$ (valence $4f$) in rare-earth magnetic materials. Following the pioneer observation of linear magnetic x-ray dichroism in $4f$ rare-earth magnetic compounds by Laan *et al.*,[4] x-ray magnetic circular dichroism in a large number of rare-earth and transition-metal systems has been reported by Schütz and coworkers who have used circularly polarized synchrotron hard x-rays,[5] and by Kao *et al.* who have used linearly polarized soft x-rays to observe resonant magnetic exchange scattering at the $L_{2,3}$ edges in Fe.[6]

In this paper we report the observation of a strong MCD at the $L_{2,3}$ edges of ferromagnetic nickel. The measurements were performed by means of a straightforward adaptation of our Dragon monochromator at the National Synchrotron Light Source.[7] The results are qualitatively consistent with a simple exchange-split-valence-band model.[3] Quantitatively, however, the intensity ratios of the L_2 and L_3 white lines and their MCD counterparts do not agree with this simple model, suggesting that the spin-dependent density of states near the Fermi level is substantially modified by the LS coupling and the Zeeman effect. This, as well as the observation of fine structures in the MCD spectra, indicates the sensitivity and usefulness of soft-x-ray MCD in the study of the electronic structure of magnetic materials.

The beamline optical arrangement is illustrated in Fig. 1(a). The decoupled horizontal and vertical focusing mirrors (HFM, VFM) of the Dragon beamline allow us to select any center for the vertical angular acceptance within a range $\psi = 4$ mrad without affecting the horizontal focusing of the monochromator. By translating the VFM vertically, one can position the center of radiation at a specific offset angle, ψ_{off}, with respect to the orbit plane. To restore full illumination of the grating, the entrance slit must then be translated vertically in proportion to the translation of the VFM. For any given ψ_{off}, the grating scanning angle and exit-slit distance were recalculated to take into account the change in the grating-incidence angle. The data presented here were taken with 12 mrad horizontal angular acceptance and 0.44 mrad vertical angular acceptance ($\Delta\psi$).

The Ni $L_{2,3}$ photoabsorption spectra were taken at normal incidence on a Ni(111) single crystal by monitoring the sample photocurrent. As shown in Fig. 1(b), the sample was rigidly attached to a view port at the back of a small vacuum chamber. In this way, the sample was electrically isolated and its surface was only a short distance ($\sim$4 mm) from a permanent magnet held outside the chamber. The magnetic field at the surface of the sample was measured to be $\sim$2000 G, a value sufficient to fully align the bulk magnetic domains at room temperature. The parallel or antiparallel orientation between the spin direction of the photons and that of the majority $3d$ electrons was selected by simply reversing the orientation of the external permanent magnet. Two metal rings, positioned in front of the sample, were biased to prevent the

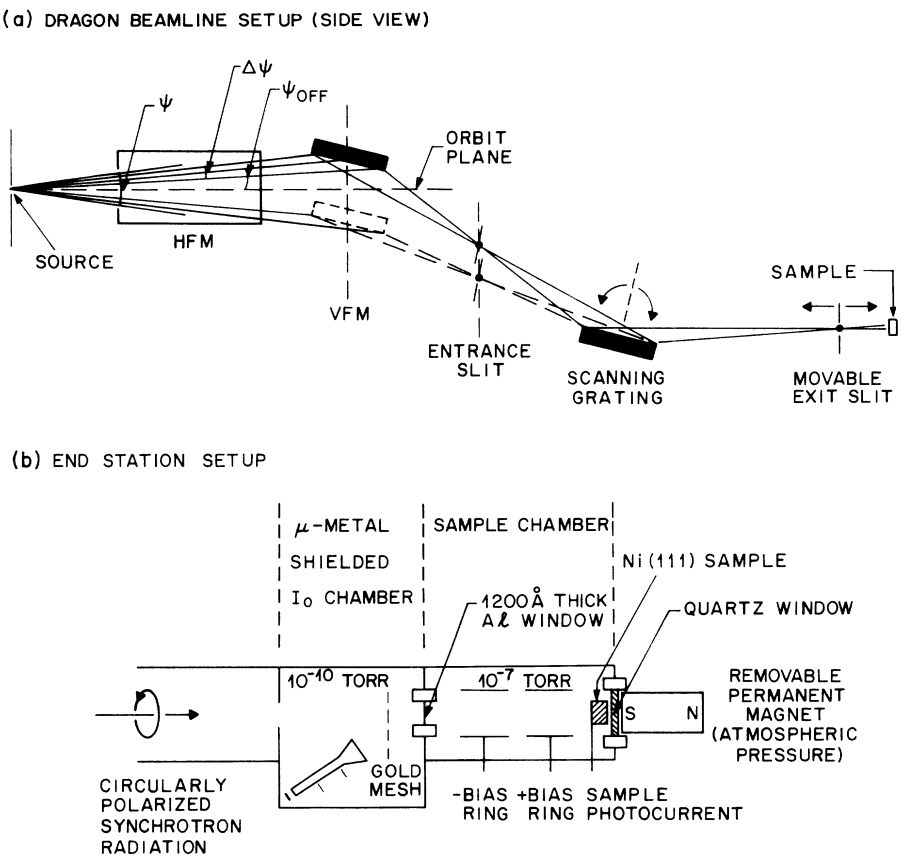

FIG. 1. (a) Beamline optical arrangement for obtaining circularly polarized soft x-rays. (b) Experimental setup for measuring the magnetic circular dichroism in nickel.

sample from recapturing the photoemitted electrons as well as from collecting electrons generated at the aluminum window. During the absorption measurements, the I_0 beam intensity was continuously monitored by detecting the total electron yield of a 90% transmission gold mesh, mounted in a μ-metal-shielded I_0 chamber [see Fig. 1(b)].

With this setup, no observable influence of the permanent magnet was observed in either the sample or the I_0 signals. Our measurements on the $L_{2,3}$ edges of nonmagnetic Ti and Cu samples at any ψ_{off} and on magnetic Ni samples at $\psi_{\text{off}}=0$ all showed the same signal readings regardless of the location and orientation of the magnet. When taking data for a given ψ_{off}, a single-sweep spectrum was taken alternately for the two different magnet orientations to minimize any possible time-dependent drift in our apparatus. The data-acquisition time for a single sweep was 9 min, and more than 15 spectra were recorded for each magnet orientation to secure the reliability of I_0 normalization and to improve the data statistical accuracy.

The $L_{2,3}$ photoabsorption spectra of the nickel crystal taken for the two different magnetization orientations are displayed in Fig. 2(a). The solid curve is the spectrum taken with the spin direction of the photons parallel to that of the majority $3d$ electrons, while the dashed curve is taken with these two spins being antiparallel to each other. In these data, taken with $\psi_{\text{off}}=0.75$ mrad above the orbit plane, the circular polarization of the monochromatized light is calculated to be $85\% \pm 5\%$. Since there is no significant MCD effect expected below and above the L_3 and L_2 white lines, the nearly perfect match of these two spectra at photon energies below 850 eV and above 880 eV demonstrates that our detection scheme is not influenced by the magnetic field, and that the I_0 normalization is very reliable.

As observed in Fig. 2(a), the intensities of the L_3 and L_2 white lines, due to essentially the $2p_{3/2} \rightarrow 3d$ and $2p_{1/2} \rightarrow 3d$ transition, respectively, are different for opposite magnetic-field orientations, and the enhancement or decrease observed at the L_3 line is reversed at the L_2 line. This effect is more visible in the difference spectrum shown in Fig. 2(b), and referred to as the MCD spectrum. By collecting radiation below the orbit plane the effect observed at the two white lines is reversed in sign, and, at any ψ_{off}, spectra taken with no magnetic field are equal to the average of those taken with opposite magnetic field.

In broad outline, our results are consistent with the simple exchange model of Erskine and Stern.[3] The essence of this model is the observation that the unoccupied $3d$ states of ferromagnetic Ni occupy a very narrow energy range just above the Fermi level and are (with neglect of spin-orbit interaction) exclusively of minority spin character ($3d\downarrow$). A straightforward application of dipole-selection

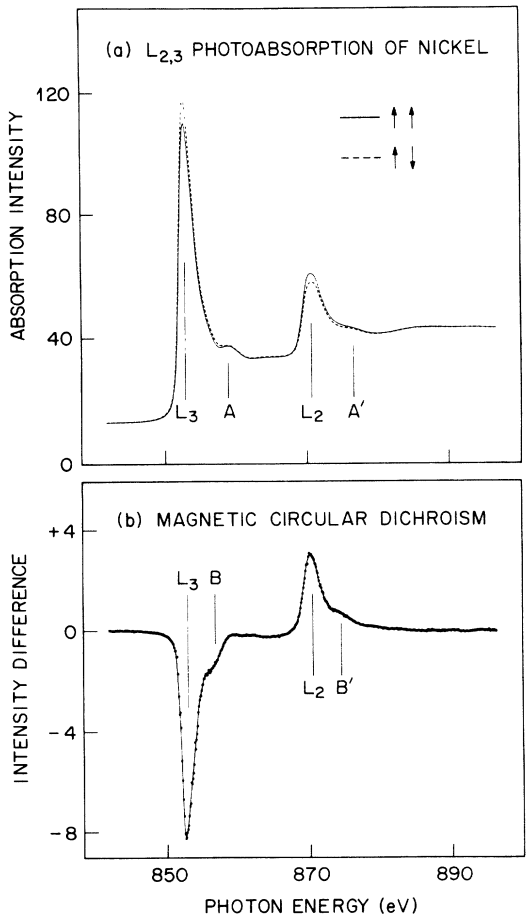

FIG. 2. (a) $L_{2,3}$ absorption spectra of nickel taken at two different orientations between the spin direction of the incident photons and that of the majority $3d$ electrons. Peaks observed at 853 and 871 eV are the L_3 and L_2 white lines. These two spectra are obtained by normalizing the Ni(111) photocurrent with the I_0 beam intensity and then scaled to make the averaged peak height of the L_3 white line equal to 100. (b) Magnetic circular dichroism of nickel obtained from the intensity difference of the two spectra shown above. The solid curve is a smoothing of the data. Fine features labeled as A,A' and B,B' are discussed in the text.

rules yields the following expressions for the total cross sections $(\sigma_+ + \sigma_-)$ and the MCD cross sections $(\sigma_+ - \sigma_-)$:

$$L_2(2p_{1/2} \rightarrow 3d\downarrow)\begin{cases}\sigma_+ + \sigma_- = \frac{2}{3}A + \frac{2}{3}B + \frac{2}{3}C\,, \\ \sigma_+ - \sigma_- = \frac{2}{3}A - \frac{2}{3}C\,,\end{cases}$$

$$L_3(2p_{3/2} \rightarrow 3d\downarrow)\begin{cases}\sigma_+ + \sigma_- = \frac{4}{3}A + \frac{4}{3}B + \frac{4}{3}C\,, \\ \sigma_+ - \sigma_- = -\frac{2}{3}A + \frac{2}{3}C\,.\end{cases}$$

The quantities A, B, and C are proportional, respectively, to the squared dipole matrix elements

$$|\langle Y_2^{\pm 2}|x \pm iy|Y_1^{\pm 1}\rangle|^2,\ |\langle Y_2^{\pm 1}|x \pm iy|Y_1^{0}\rangle|^2,$$

and

$$|\langle Y_2^{0}|x \mp iy|Y_1^{\pm 1}\rangle|^2,$$

where Y_l^m are spherical harmonics and the $x \pm iy$ is the dipole operator of circularly polarized photons. No matter how the A, B, and C terms are weighted,[8] the model predicts that the ratio of total cross sections for L_3 and L_2 white lines be 2:1, and the corresponding ratio for the MCD cross sections be -1:1. Qualitatively, these predictions are not too bad. Quantitatively, the measured peak area ratios are about 2.6:1 for the total cross sections and about -1.6:1 for the MCD cross sections. This appreciable difference could be related to the change of spin dependent unoccupied density of states near the Fermi level caused by the LS coupling and the Zeeman effect. It has been noted previously in the related case of Pt that spin-orbit interaction within the valence d band will enhance the proportion of $j = \frac{5}{2}$ character at the top of the d band and therefore favor the L_3 over the L_2 edge.[9] Thus we may conclude that the MCD measurements are sensitive to details of the electronic and magnetic structure beyond elementary atomic-dipole selection rules, and call for more sophisticated calculations of the kind which are now beginning to appear in the literature.[4,6,10]

In support of this conclusion, we note the existence of weaker features in the spectra at about 4 and 6 eV above their respective white lines. The 6 eV feature [labeled as A,A' in Fig. 2(a)] appears only in the total spectrum, and as has been noted before,[11] is attributed to an edge in the density of states starting at the onset of band 7 at the symmetry point L_1. Exchange splittings are negligible this high in the unoccupied band structure, and so it is not surprising that these features have no counterparts in the MCD spectrum. By way of contrast, the 4 eV feature [labeled as B,B' in Fig. 2(b)] is imperceptive in the total spectrum but manifests a distinct shoulder in the MCD spectrum. We tentatively attribute this feature to exchange-split d states weakly hybridizing with the unoccupied s,p states above the nominal top of the d band.

In summary, we have observed MCD at the $L_{2,3}$ edges of ferromagnetic Ni. The results are sensitive to the details of the electronic and magnetic structure, thus confirming the optimism expressed earlier by others.[3-6] The strong MCD at the $L_{2,3}$ edges of nickel, approximately 2 orders of magnitude larger than those at the K edges of transition metals,[5] motivates the further use of circularly polarized soft x-rays to study magnetically ordered $3d$ ferromagnets and $4f$ rare-earth materials.

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