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Spin-aligned momentum distributions of transition-metal ferromagnets studied with circularly polarized synchrotron radiation

Dennis M. Mills
Cornell High Energy Synchrotron Source and the School of Applied and Engineering Physics, Cornell University, Ithaca, New York 14853
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The momentum distributions of the unpaired spin electrons in Fe, Co, and Ni have been measured using circularly polarized x rays produced from an x-ray phase plate developed at the Cornell High Energy Synchrotron Source. Estimates of the negative polarization of the s-p components of the conduction electrons have been made. The spin-dependent transition-metal momentum profiles are compared to theoretical calculations. Preliminary results from gadolinium are also presented.

Measurements of the momentum distribution of the spin-aligned electrons in ferromagnetic materials have traditionally fallen into the domain of spin-polarized positron annihilation studies.[1] In 1970 Platzman and Tsoar[2] pointed out that x rays could be used for studying the momentum distribution of unpaired spin electrons through inelastic or Compton scattering. Although Compton scattering is usually considered in terms of the interaction between the incident x ray and the *charge* of the electron, if the incident x-ray beam is elliptically polarized, the scattering from the electron's *spin* can be experimentally separated from the charge scattering. Using circularly polarized γ rays emitted by a cooled ^{57}Co source Sakai and Ono[3] in 1976 measured the Compton profile of the magnetic electrons in iron. This formidable experiment was limited to a source strength of only 10 mCi (due to self-heating) and therefore nearly 140 h of data collection was required to accumulate a magnetic Compton profile. In order to make magnetic scattering a more useful tool, high-flux sources of elliptically polarized x rays need to become available. Synchrotron radiation can provide such a source. Recently Cooper *et al.*[4] have used that portion of the synchrotron radiation that is naturally elliptically polarized to remeasure the magnetic Compton profile in iron. Unfortunately, only a small fraction of the total power emitted from a storage ring source is naturally circularly polarized. This significant reduction of intensity coupled with the experimental problems that must be handled in order to make use of the elliptically polarized x rays make such experiments nonroutine. To avoid these difficulties, the circularly polarized x rays used in this experiment were produced from the predominantly linear polarized synchrotron radiation beam by means of a perfect double-crystal monolithic monochromator with one of the crystals arranged in the Laue orientation and acting as an x-ray phase plate.[5]

The theoretical framework for understanding Compton scattering of circularly polarized x rays from the electron's spin has been discussed by Lipps and Tolhoek[6] and Platzman and Tzoar.[2] Following their notation, the Compton cross section (including the spin contribution) can be written as

$$\frac{d^2\sigma}{d\Omega\, dp_z} = \frac{r_0^2}{2}\left(\frac{k_s}{k_0}\right)^2 [(\Phi_0 + P_l\Phi_1)J^+(p_z) + P_c\Phi_2(\boldsymbol{\sigma})J^-(p_z)] ,$$

where r_0 is the classical radius of the electron, $\mathbf{k}_0(\mathbf{k}_s)$ the incident (scattered) x-ray wave vector, and $P_l(P_c)$ is the percent linear (circular) polarization of the incident beam. The angular distribution of the scattered radiation is contained in the Φ_i's:

$$\Phi_0 = (1+\cos^2\phi) + (k_0 - k_s)\frac{\hbar c}{m_0 c^2}(1-\cos\phi) ;$$

$$\Phi_1 = \sin^2\phi;\ \Phi_2(\boldsymbol{\sigma}) = -(1-\cos\phi)\boldsymbol{\sigma}\cdot(\mathbf{k}_0\cos\phi + \mathbf{k}_s)\frac{\hbar c}{m_0 c^2} .$$

Here, $\boldsymbol{\sigma}$ is the electron's spin, ϕ the scattering angle, and m_0 the rest mass of the electron. Information on the shape of the Compton profiles is contained in the $J(p_z)$'s, which are the integrals over p_x and p_y of the sum or difference of the spin-up $[n^{\uparrow}(\mathbf{p})]$ and spin-down $[n^{\downarrow}(\mathbf{p})]$

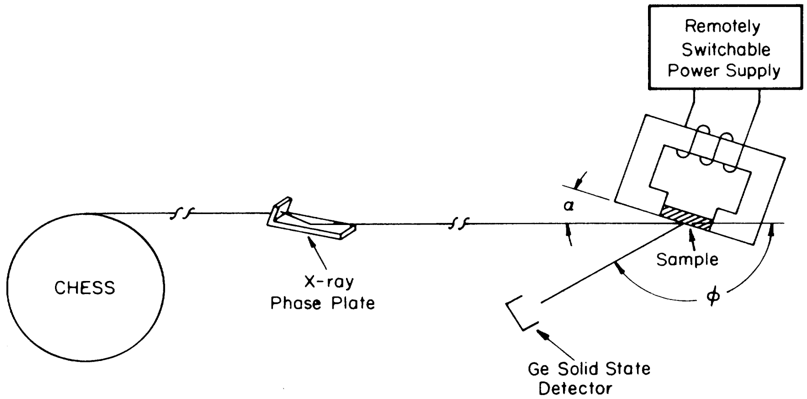

FIG. 1. Schematic of the experimental geometry. The synchrotron radiation emitted from CHESS was altered from linear to circular polarization via the x-ray phase plate. This beam was then incident on the sample that was positioned between the poles of a remotely controllable electromagnet. The Compton scattered radiation was recorded by an intrinsic germanium solid-state detector.

momenta distributions, i.e.,

$$J^{\pm}(p_z)=\int\int[n^{\uparrow}(\mathbf{p})\pm n^{\downarrow}(\mathbf{p})]dp_x\,dp_y\ .$$

We see that only $\Phi_2(\boldsymbol{\sigma})$ is spin dependent and that this term is a function of both the magnitude and direction of the spin. This makes possible the elimination of the charge scattering component (leaving only the spin contribution) by making measurements at two different spin directions. Experimentally this can be realized when working with ferromagnetic samples by the application of an external magnetic field. A cross-section difference (spin up minus spin down) can be written as

$$\left(\frac{d^2\sigma}{d\Omega dp_z}\right)_{\Delta}=P_c r_0^2\left(\frac{k_s}{k_0}\right)^2\Phi_2(\sigma)J^{-}(p_z)\ .$$

As can be seen from above, the spin term is nonvanishing only if the incoming beam has a component of circular polarization.

The experimental geometry used for these measurements is shown in Fig. 1. The samples were placed across the poles of an electromagnet whose field direction could be reversed. The angle α between the incident wave vector $\mathbf{k}_0$ and the direction of the spin alignment $\boldsymbol{\sigma}$ was approximately 10–15°. The Compton scattering profile was measured at a scattering angle ϕ of $\sim 150°$ with a germanium solid-state detector. $\boldsymbol{\sigma}$, $\mathbf{k}_0$, and $\mathbf{k}_s$ were all coplanar.

The magnetic field was reversed and hence data collected in an *ABBA* sequence. *BAAB* switching produced identical results. One *A* or *B* step was approximately 2 min in length with many sequences summed to produce the data shown below. The iron data required about 14 h of counting time at Cornell High Energy Synchrotron Source (CHESS) with the storage ring running at 5.5 GeV and 25 mA. All data were recorded with polycrystalline samples at room temperature with the exception of the gadolinium data which were recorded at $-40\,°C$. Figure 2 shows the raw spectrum for iron in one spin orientation. The incident x-ray energy was about 40 keV, and with a scattering angle 150° the Compton-shifted profile is centered around 35.2 kev. The width of the elastic line is due solely to the detector resolution ($\Delta E = 500$ eV at 40 keV).

The momentum distributions of the unpaired spin electrons in iron, cobalt, and nickel are shown in Fig. 3. The ordinate of the data has been transformed from energy to momentum in atomic units (a.u.), and folded about $p_z = 0$. No corrections were made for multiple-scattering effects. Data were also taken in the identical geometry with nonmagnetic samples (Al and Cu) to rule out possible biasing as a function of the magnetic field direction. The accuracy of the subtraction process can be gauged by observing the residual of the elastic peak that occurs near $p_z = 12$ a.u.

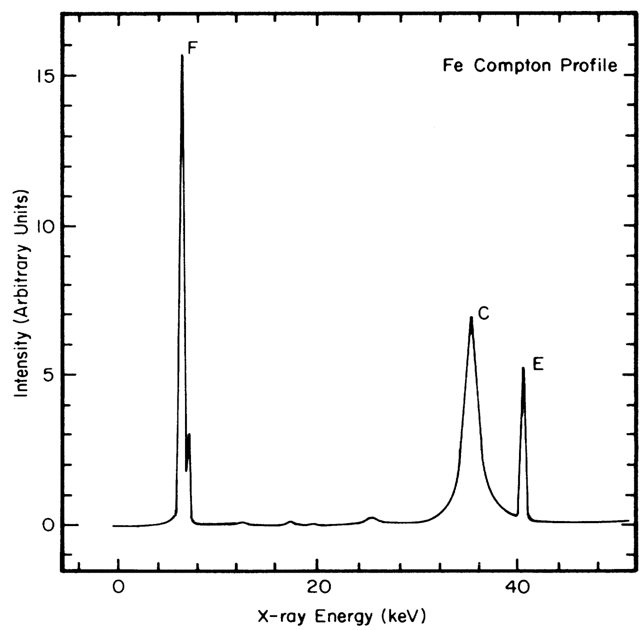

FIG. 2. Raw data from the solid-state detector for one spin orientation of iron. The large peak near 6 keV is Fe *K* fluorescence. The Compton and elastically scattered peaks are labeled C and E, respectively.

The solid lines in the iron and nickel data are calculated profiles obtained from spin-polarized band structures determined by augmented plane wave[7] and orthogonal plane wave plus linear combination at atomic orbitals[8] for iron and nickel, respectively. The calculated profiles have been convoluted with the resolution function of our experiment (~1 a.u.). The experimental data were normalized to the calculations by equating the areas under the curves from 0 to 6 a.u. Although the general agreement between the calculations and experimental data is good, both calculations tend to underestimate the component of the unpaired spin electrons that want to align in a direction opposite that of the majority electrons (negative polarization component), which manifests itself in the Compton profile as a dip about $p_z = 0$. (Physically this corresponds to a concentration of negative magnetization density at the interatomic regions of the crystal lattice.) Because the integral over p_z of the Compton profile is equal to the net magnetic moment of the atom in Bohr magnetons (μ_B) one can also express the magnitude of the negative polarization component in these units. Negative polarizations of $(-0.20 \pm 0.05)\mu_B$ and $(-0.07 \pm 0.05)\mu_B$ were experimentally determined for Fe and Ni, respectively. The calculations by Wakoh and Kubo[7] for iron gave a value of $-0.07\mu_B$ and those by Rennert, Carl, and Hergert[8] for nickel a value of $-0.02\mu_B$. Calculations for the magnetic Compton profile in cobalt could not be found. The negative polarization in cobalt was estimated to be $(-0.1 \pm 0.06)\mu_B$ from this work. The componet of negative polarization in these materials has also been determined by neutron-diffraction experiments to be -0.21 (Ref. 9), -0.28 (Ref. 10), and -0.105 (Ref. 11) Bohr magnetons for Fe, Co, and Ni, respectively. The origin of the negative polarization has been suggested to arise from the 4s conduction electrons.[9,11,12] Theoretical calcula-

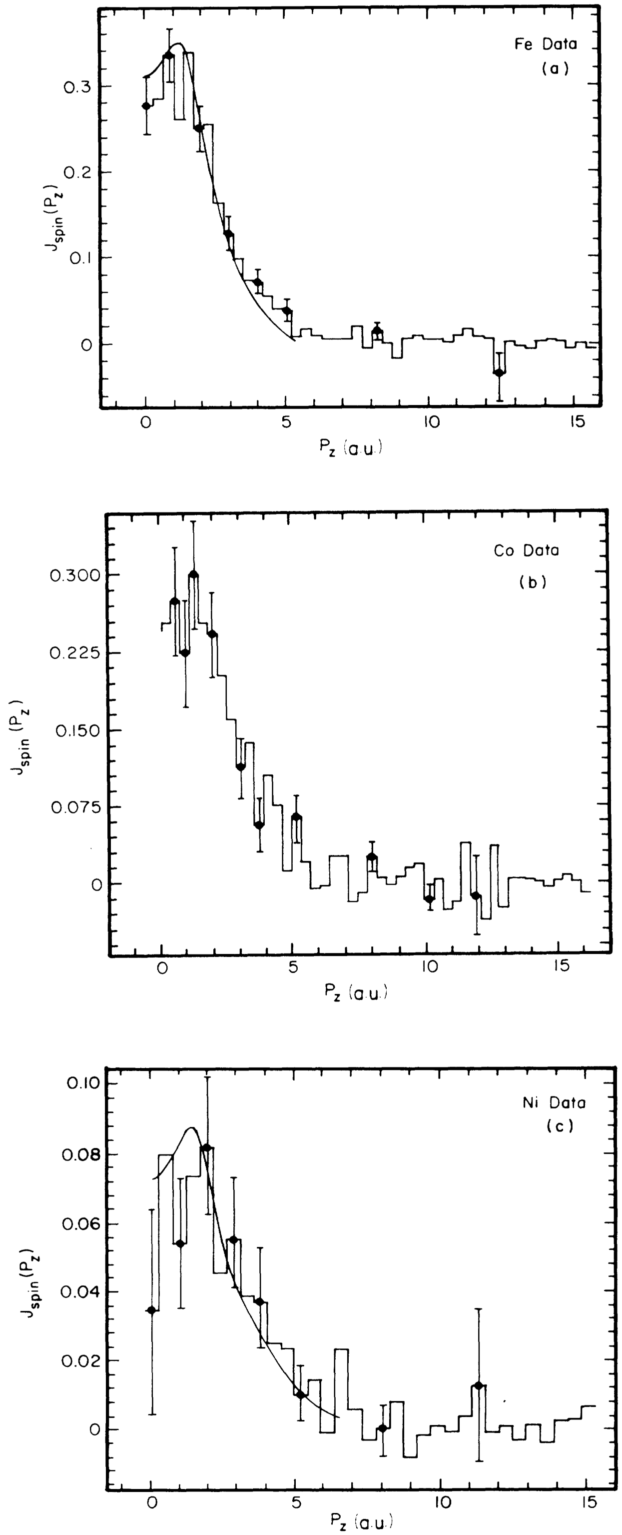

FIG. 3. The spin momentum distributions plotted as a function of momentum for (a) iron, (b) cobalt, and (c) nickel. The solid lines in the iron and nickel data are calculated values of the momentum distributions of the unpaired electrons in these materials. In each of these transition metals a clear dip centered about $p_z = 0$ is observed.

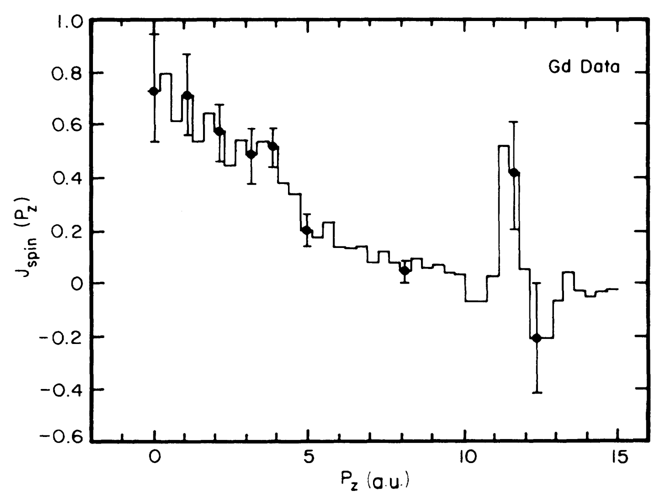

FIG. 4. The spin momentum distribution of gadolinium plotted as a function of p_z. These data were recorded at $T = -40\,°C$. The minimum at $p_z = 0$ is not observed in this case and the extent of the spin-dependent scattering is much more extended in Gd, as compared to the transition ferromagnets indicative of the fact that the origin of the magnetism is more atomiclike.

tions by Wakoh and Kubo supported this, showing that the negative polarization does in fact arise from the *s*-*p* componets of the conduction-band electrons.

Figure 4 shows the magnetic Compton profile for gadolinium which has a Curie temperature of 293 K. Although the spin magnetic momentum in gadolinium is large ($\sim 7\mu_B$) compared to the transition-metal ferromagnets $[(2.2-0.6)\mu_B]$, the total Compton scattering is considerably reduced for higher-Z materials resulting in somewhat larger error bars in the Gd data. Nonetheless, several differences in the magnetic Compton profiles can still be observed between the transition metals and rare-earth sample. The Gd data show no dip in the profile about $p_z = 0$ and the profile extends out well past 8 a.u., whereas in the transition metals there is little intensity past 5 a.u. The wider profile is the signature of atomiclike magnetism in gadolinium where the spin electrons are contained in the close-in $4f$ shells as compared to the itinerate $3d$ spin electrons in the transition ferromagnets.

In summary, the magnetic Compton profiles of three transition-metal ferromagnets were measured using circularly polarized x rays produced with an x-ray phase plate. The general shape of the profiles were similar, each having a characteristic dip near $p_z = 0$ and extending to about 5 a.u. A measure of the negative polarization componet for Fe, Co, and Ni was found to be -0.20 ± 0.05, -0.10 ± 0.06, and $\pm 0.07 + 0.05\ \mu_B$, respectively. The Gd magnetic Compton profile was also measured at $-40\,^\circ$C. No dip at $p_z = 0$ was observed and the profile extended out to much larger values of momentum, indicating the atomiclike origin of the magnetization experiments described here were recorded at CHESS with radiation from one of the dipole magnets ($E_{\mathrm{crit}} = 10$ keV). Recent additions to the CHESS facility now permit user access to the white beam from a six-pole electromagnetic wiggler, which under the identical machine parameters that these data were recorded would produce a factor of 60 increase in flux at 40 keV. This, coupled with the increased currents (> 65 mA) should allow one to extend these type of measurements to better resolution and to look at single-crystal samples in order to observe directional Compton profiles in the very near future.

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