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Spin-dependent momentum distribution in iron studied with circularly polarized synchrotron radiation

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The spin-dependent Compton profile of ferromagnetic iron has been measured with circularly polarized synchrotron radiation extracted at 46 and 62 keV from the Daresbury storage ring by viewing radiation from the 5-T magnet at 0.24 mrad above and below the electron orbital plane. For the first time (to our knowledge) in a photon scattering experiment the line shape is well established. The negative magnetization resulting from s-p polarization is evident here as a 25% dip around zero momentum, which is greater than that predicted by an augmented-plane-wave calculation of this effect.

In a recent paper we reported the first measurement of the spin-dependent Compton profile of iron with circularly polarized synchrotron radiation.[1] The method, proposed earlier by two of us,[2] for the extraction of the circularly polarized component was to view the source at a small angle to the orbital plane at the tangent point of a bending magnet. That experiment was successful as a demonstration of the principle, but the photon flux was lower than predicted and the statistical accuracy insufficient to permit any sensible interpretation of the line shape. The present study, at higher photon energies and with a different monochromator, has now yielded fluxes in line with predictions. The Compton profiles have far better statistical accuracy and can be directly interpreted in terms of the negative s-p polarization of the conduction electrons. These results should provide a spur to the further study of several x-ray inelastic magnetic scattering effects which have been discussed recently elsewhere.[3,4]

The cross section for right-hand-circularly-polarized γ rays of energy ω_0, scattered by a free electron with spin parallel (+) or antiparallel (−) to the incident photon's wave vector is given as[5]

$$\left(\frac{d\sigma}{d\Omega}\right)^{\pm}=\frac{1}{2}\left(\frac{e^2}{mc^2}\right)^2\left(\frac{\omega}{\omega_0}\right)^2\left[\left(\frac{\omega_0}{\omega}+\frac{\omega}{\omega_0}-\sin^2\phi\right)\pm\left(\frac{\omega_0}{\omega}-\frac{\omega}{\omega_0}\right)\cos\phi\right], \quad (1)$$

where ϕ is the angle of scattering and ω is the energy of the scattered photon; ω_0 and ω are related by the equation $\omega_0/\omega=1+\omega_0/mc^2(1-\cos\phi)$. The last term, which is of order ω_0/mc^2 is only present if circularly-polarized radiation is used and it changes sign with the hand of the radiation.

In reality the target electrons are moving and this leads to a spectral distribution, $d^2\sigma/d\Omega\, d\omega$, for the scattered photons which is related to the momentum distribution $n(\mathbf{p})$ of those electrons by the Compton profile integral[6]

$$J(p_z)=\int\int n(\mathbf{p})dp_x dp_y \ , \quad (2)$$

i.e., it is the projection of the momentum distribution along the scattering vector (chosen as the z direction) and all electrons contribute to the integral.

In the case of a system of aligned spins $\boldsymbol{\sigma}$ the spin-dependent contribution can be isolated by reversing the photon polarization or the magnetization direction to yield a cross section involving the momentum distribution of the unpaired spins,

$$\Delta\frac{d^2\sigma}{d\Omega\, d\omega}=\left(\frac{d^2\omega}{d\Omega\, d\omega}\right)_{\text{up}}-\left(\frac{d^2\sigma}{d\Omega\, d\omega}\right)_{\text{down}}=\left(\frac{e^2}{mc^2}\right)^2 P_c\boldsymbol{\sigma}\cdot\mathbf{C}J_{\text{mag}}(p_z) \ , \quad (3)$$

where P_c is the degree of circular polarization and $\mathbf{C}$ is a vector of magnitude ω_0/mc^2, the exact value of which depends upon the scattering geometry.[7] The magnetic Compton profile, $J_{\text{mag}}(p_z)$, is written as

$$J_{mag}(p_z)=\int\int\sum_i[n_{i,\text{up}}(\mathbf{p})-n_{i,\text{down}}(\mathbf{p})]dp_x dp_y \ , \quad (4)$$

where $n_i(\mathbf{p})$ is the "up" or "down" momentum density in the ith band and the sum is over all bands. It is more structured than the total profile because of the major cancellation of the spin-paired electron contributions, and this is clearly demonstrated in iron in the magnetic profile shown in Fig. 1 which was calculated by the augmented-plane-wave (APW) method.[8] The $3d$-electron distribution is relatively flat at momenta below 2 a.u., but there is a pronounced minimum—some 15% of the peak height—around the origin. This is a consequence of the negative s-p polarization which also manifests itself in position space as negative magnetization density in the interatomic regions. The aim of this experiment was to achieve sufficient accuracy and resolution to provide a quantitative check on this structure in the magnetic profile.

The first measurement of the magnetic profile of iron was made one decade ago[9,10] using nuclear orientation in ^{57}Co to achieve an appreciable degree of circular polarization in the 122-keV γ-ray emission. The source strength

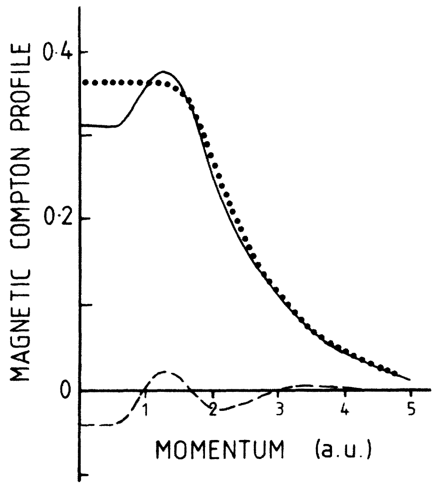

FIG. 1. Partial contributions to the magnetic Compton profile of iron calculated according to the APW method by Wakoh and Kubo (Ref. 8). The dotted line represents the $3d$ contribution and the dashed line the s-p components. The solid line is the total. A small contribution from the polarization of the core has been neglected.

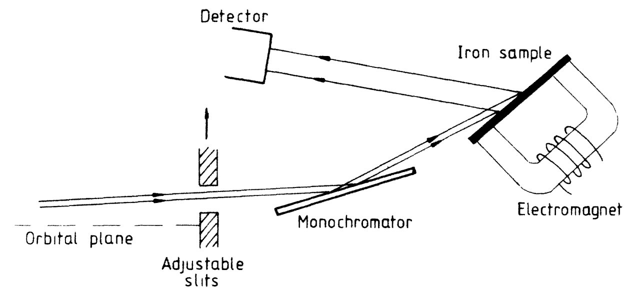

FIG. 2. Schematic diagram of the apparatus used to detect magnetic Compton scattering. The beam is selected by a series of slits (one set shown) at an angle 0.24 mrad to the orbital plane of the storage ring. The radiation from the tangent point of the 5-T wiggler magnet is monochromated with a Ge{111} crystal, and the diffracted beam impinges upon the soft iron sample at a small angle of incidence. The magnetization cycle is controlled by a microcomputer and the signal from the germanium semiconductor detector routed by the computer into alternate 2048 channel memories.

was only 10 mCi because self-heating imposes an upper limit if the temperature is to be low enough (e.g., below 50 mK) to produce an appreciable degree of circular polarization. In that experiment only 4×10^4 counts were accumulated in the difference profile in the measurement time of ~140 h. A dip of approximately three times that predicted ($\sim50\%$ of the peak height) was apparent in the data, but the statistical accuracy was too low to estblish its significance. In a more recent experiment[11] a 40-mCi ^{191}Os source ($E_\gamma=129$ keV) yielded increased flux ($\sim6\times10^5$ counts in 165 h), but in the experiment there was less evidence of a central dip in the magnetic Compton profile.

The method used here relies on the fact that the synchrotron radiation emerging from the tangent point of a dipole bending magnet is elliptically polarized above and below the machine's orbital plane. At photon energies between 40 and 60 keV an angle of $\frac{1}{4}$ mrad is sufficient to produce a beam with $P_c>50\%$, but at this angle the intensity is already two orders of magnitude lower than in the orbital plane.[2] In this experiment the three-pole wiggler magnet port on the Daresbury Synchrotron Radiation Source (SRS) was used. In our previous study[1] an adjacent station which may have inadvertently afforded a view of the 2.5-T upstream magnet[7] as well as the central 5-T "wiggle" was used. This may have led to some polarization cancellation[12] because of the reversal of the beam curvature and hence, the hand of the circular polarization. In this experiment no such cancellation should occur.

The apparatus is outlined in Fig. 2. The saturation magnetic field was imposed on the soft iron sample by an electromagnet, the field being reversed at 10- and 20-s intervals in an asynchronous cycle of period 80 s and the data switched between two memory stores at each reversal. After an initial experiment "on orbit" which yielded the anticipated null result (because $P_c=0$) the apparatus was moved up 8 mm in the vertical plane to collect radiation emerging at 0.24 mrad from the source some 35 m distant. A slit of size 7 mm wide $\times5$ mm high was used to define the beam onto the germanium single-crystal monochromator which was oriented to select an energy aroung 45 keV with the (333) Bragg reflection. The scattering angle was $151°\pm2°$. The SRS operates on 8-h cycles so each run was timed for 6 h and resulted in 2×10^7 counts in both up and down profiles. A typical spectrum is shown in Fig. 3(a), where Compton profiles from both 46.4-keV [Ge (333) reflection] and 61.9-keV [Ge (444) reflection] incident beams are clearly visible, together with the narrower elastically scattered lines.

Figure 3(b) shows the difference profile accumulated from two such runs; these data were collected in two 8-h shifts. The elastic lines have canceled, because at $\sin\theta/\lambda\cong4$ Å^{-1} only the spin-paired core electrons have a measurable x-ray scattering factor, but the magnetic Compton profiles from both incident beams are clearly visible. The (333) difference profile contains 3.6×10^5 counts and amounts to 2% of the unpolarized peak, in line with our calculations for this energy. The anticipated dip in the magnetic Compton profile (see Fig. 1) is actually visible in the raw data at both energies. Around 60 keV, where the difference spectrum should be zero there is a small contribution from another magnetic Compton profile. This arises from 77-keV radiation diffracted by the (555) reflection of germanium. It is approximately 10% of the (444) magnetic profile, in line with calculation. Additional measurements were made as further checks: first, one 8 mm below the orbit and second, one with the magnetic field cycle reversed. Each change inverted the magnetic Compton profile.

The data were corrected for the energy variation of both the Compton cross section and the sample absorption. They were then converted to a momentum scale and the profile normalized in the range $|P_z|<5.0$ a.u. to the same area as the theory. No corrections for multiple scattering were made. The resulting profiles for both

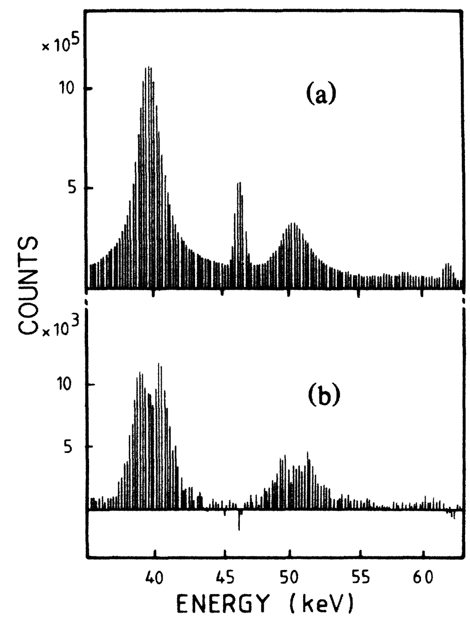

FIG. 3. Experimental Compton profiles of iron: (a) the total profile and (b) the magnetic (difference) profile. Radiation at 46.4 and 61.9 keV is selected by the (333) and (444) reflections of the germanium monochromator, and the Compton profiles corresponding to the 151° ± 2° scattering angle are at 39.7 and 50.4 keV, respectively. The count refers to a 100-eV energy bin and was accumulated over a 12-h measurement period. The difference profile is essentially zero at the elastic line positions but the magnetic profiles and their "volcano" structure are readily evident.

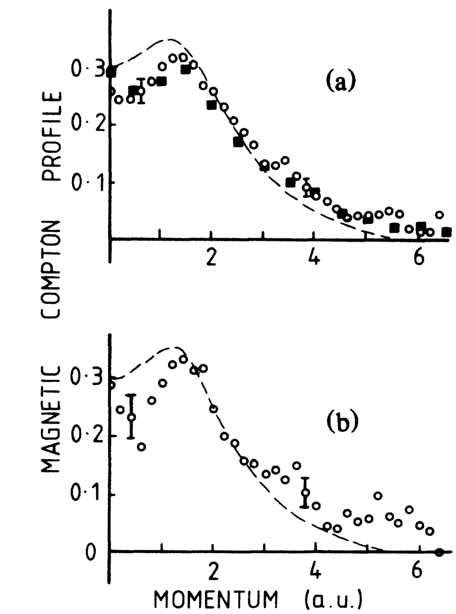

FIG. 4. Processed data for (a) low-energy and (b) high-energy magnetic profiles compared with theory. The data have been averaged from left to right and normalized to the same area as the theory. The present synchrotron data are shown as open circles; the filled squares in (a) represent the radioisotope data (Ref. 11) which were reported at the same resolution and similar statistical accuracy. The APW curve (Ref. 6) has been convoluted with Gaussians of (a) 1.0 a.u. FWHM and (b) 0.8 a.u. FWHM to facilitate the comparison with experiment.

(333) and (444) profiles were symmetrical, as they should be [$n(\mathbf{p})$ is a centrosymmetrical function] and were therefore averaged from left to right to yield the final result displayed in Fig. 4.

The standard deviation, per 0.2-a.u. momentum interval is approximately one quarter of the predicted dip and therefore adequate to characterize this feature. The energy resolution of the germanium detector limited the profile resolution to 1.0 a.u. full width at half maximum (FWHM) at the lower energy and 0.8 a.u. at the higher. For comparison the APW theory was convoluted with Gaussians of these widths. This resolution is rather worse than in conventional Compton scattering experiments. However, the magnetic profile is approximately twice the width of the all-electron profile and the limited resolution is therefore not a problem.

Referring to Fig. 4 it is clear that this experiment verifies the existence of a central dip in the magnetic profile and that its depth is approximately twice the prediction of the APW model; its width is correctly predicted. According to Platzman and Tzoar[4] the depth of the minimum is a measure of the negative spin polarization and the degree of s-d exchange interaction.

Both experimental profiles extend further than the theoretical ones, with significantly more momentum density beyond 4 a.u. There appears to be some evidence for this in the earlier results.[9,10] Although Sakai, Terashima, and Sekizawa[11] did not attempt any interpretation of the line shape, we have averaged their result from left to right and included it in Fig. 4(a) for comparison. The central dip is not well established in their data but the two distributions are indistinguishible at higher momenta. If this extra density at high momenta is real it indicates that the plateaulike d-electron contribution (see Fig. 1) extends further out in momentum space than predicted, i.e., more d components with positively polarized spins. It is possible that the APW calculation does not include enough of the high-momentum component of the $3d$ wave functions. The momentum density is synthesized from an expansion of eigenfunctions over a limited range of reciprocal-lattice vectors, i.e., the higher-order terms are excluded. Nor does this discrepancy contradict the agreement between this theory and earlier spin-polarized positron measurements:[13,14] The positron is excluded from the positively charged iron core and is therefore less sensitive than the photon to the high-momentum components of the wave function.

It is hoped that this experiment will be repeated at higher primary energies with improved statistical accuracy and resolution, in order to pinpoint these effects more precisely.

In conclusion, we note that these measurements were not primarily limited by count-rate considerations, in contrast to the radioisotope measurements. Blume[3] has pointed to the wealth of information about magnetic systems embedded in the x-ray scattering cross section. Even without tailor-made polarization devices the coupling between circularly polarized radiation and unpaired spin electrons can be exploited to yield information about spin density complementary to that produced by neutron scattering, as has been illustrated in this study. An order of magnitude gain in signal, coupled with improvements in resolution, is possible even with the present method. The implications for magnetic scattering studies on the next generation synchrotrons are profound.

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