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LETTER TO THE EDITOR

Does magnetic Compton scattering only measure spin magnetization?

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Abstract. Experimental results with circularly polarized synchrotron radiation show that magnetic Compton scattering arises solely from the spin magnetization in the sample. This observation disagrees with the cross-section recently derived for bound electrons. The measurements were performed on $HoFe_2$, a ferrimagnet with dominant orbital magnetization.

The question of whether inelastic, incoherent scattering experiments can be used to separate electron spin and orbital contributions to magnetization became of great interest following experimental evidence published by some of the present authors [1] supporting such a theoretical prediction [2]. This letter reports a more incisive measurement showing that no significant orbital contribution is present in magnetic Compton scattering under the conditions usually associated with these experiments.

The opportunity to study magnetization densities by x-ray as well as neutron diffraction techniques has arisen with the development of synchrotron sources capable of producing high-brightness x-ray beams with well defined polarization states. The cross-section for elastic scattering, which has been derived by a number of authors [3, 4] contains terms relating to both spin and orbital magnetization. The formulae are confirmed by number of diffraction experiments [5, 6]. In the case of ferromagnets, where charge and magnetic Bragg peaks are always superimposed, measurable effects, at the energies away from absorption edges, only occur if circularly polarized photons are used. This produces an interference term in the cross-section which contains separate contributions from F_S and F_L, the spin and orbital magnetization per unit cell, which leads to the intriguing possibility of directly determining spin and orbital magnetization separately.

Compton scattering, which is both inelastic and incoherent, can only be used to study magnetism in materials that have a net moment. It offers some advantages over diffraction studies in that the scattering angle and the photon energy are independent parameters; the geometrical requirements are relaxed and single-crystal material is not essential. Traditionally it has been the Compton line-shape, which is a one-dimensional projection of the electron momentum distribution, that has been studied

[7]. The cross-section for magnetic Compton scattering was first calculated for a free stationary spin-polarized electron by several authors [8] and this expression was developed, within the spirit of the impulse approximation, to describe the scattering from a free target electron moving with a momentum distribution dictated by the atomic potential [9]. In this approach the question of orbital magnetization did not arise. In addition, for reasons associated with maximizing the resolution and ensuring the validity of the impulse approximation, backscattering geometry has always been adopted and in this geometry the orbital term, predicted by equation (1) below, would have been negligible. Therefore all the experimental studies of magnetic Compton profiles have been associated solely with studies of the spin-dependent distribution. They have, in any case, been performed on soft, spin-dominated ferromagnets because of the need to use rapid reversal of the sample's magnetization as a means of both separating the magnetic contribution and minimizing the effects of beam fluctuations.

The spin-dependent Compton scattering from bound electrons was calculated by perturbation theory [10]; the orbital contribution, which appears in the second-order perturbation terms [11] has been calculated more recently [2]. It can be expressed (apart from constants) in the form of two independent geometrical terms associated with $\boldsymbol{F}_S$ and $\boldsymbol{F}_L$:

$$(E/mc^2)P_c[\boldsymbol{F}_S\xi(\theta,\alpha,\varepsilon)+\boldsymbol{F}_L\zeta(\theta,\alpha,\varepsilon)] \tag{1}$$

where

$$\xi(\theta,\alpha,\varepsilon)=(1-\cos\theta)(2\cos\theta\cos\alpha+\sin\theta\sin\alpha)+\frac{\varepsilon}{2}[\cos\alpha(1+3\cos\theta)(1-\cos\theta) + \sin\theta\sin\alpha(1-3\cos\theta)] \tag{2}$$

$$\zeta(\theta,\alpha,\varepsilon)=\left[\frac{\sin\theta(1+\cos\theta)}{2(2+\varepsilon)}\right]\left[4\sin\frac{\theta}{2}\cos\left(\alpha-\frac{\theta}{2}\right) \times\varepsilon[\sin\alpha(3+\varepsilon)\sin(\theta-\alpha)]\right]. \tag{3}$$

The nomenclature used in [1] and [2] is adopted here. E and P_c are the energy and degree of circular polarization of the incident photons, θ is the scattering angle, α is an angle between the magnetization direction and the incident photon wavevector and mc^2 is the electron rest mass. The interference contribution to the cross-section has the similar terms as magnetic diffraction plus additional terms in $\varepsilon = \Delta E/E$, where ΔE is the energy transfer in Compton scattering.

This prediction for magnetic scattering in the Compton limit has led to a series of experiments aimed at verifying the existence of both spin and orbital terms in the observed scattering. The first of these were carried out on the relatively soft ferromagnets Fe and Co, which have $\boldsymbol{F}_L$:$\boldsymbol{F}_S$ ratios of 0.046 and 0.094 respectively [1]. The method involved measuring the total intensity of the magnetic Compton scattering as a function of the angle, α. With scattering angles near 100° the spin term changes sign with respect to the charge scattering at an angle α^* in the range 10–20° but the orbital term does not. Thus the exact zero-crossing angle acts as a test of the $\boldsymbol{F}_L$:$\boldsymbol{F}_S$ ratio. In practice the angular scale for α and θ could not be established with sufficient accuracy for absolute measurements; instead the change in α^* between Fe and Co was interpreted as evidence of a change in the $\boldsymbol{F}_L$:$\boldsymbol{F}_S$ ratio. The result (a shift in α^* of 2.9°) was in the sense predicted by the theory and just significant at the 3σ level; this was taken as evidence in support of the cross-section for bound electrons derived by Lovesey [2]. However lower quality data on $HoFe_2$, which has a $\boldsymbol{F}_L$:$\boldsymbol{F}_S$ ratio predicted to be approximately −3:1 at room temperature [12], but which is more difficult to saturate magnetically, followed the spin-only prediction [1, 13].

In the last two years those measurements have been repeated, with better-defined geometries, a number of times at the SRS, Daresbury in the UK and the Accumulation Ring (AR) synchrotron source at KEK, Japan by the present authors. The results, which will be described and analysed in detail elsewhere, fail to confirm the difference found in that first study. We have recently been able to carry out a much more decisive experiment to resolve this issue and the purpose of this letter is to report it briefly.

The measurements were carried out at the AR elliptical wiggler station at KEK at room temperature. The insertion device is capable of producing 'on-axis' circularly polarized radiation at energies up to 70 keV [14, 15]. The inherently higher flux, as compared to the SRS wiggler line, allowed us to use a small (2 mm × 1.5 mm × 1 mm) polycrystalline $HoFe_2$ sample. The specimen was made from high-purity materials to minimize the contamination of the Compton spectra with rare-earth K-shell fluorescent lines. The incident beam energy was deliberately chosen to be 48 keV, i.e. below the Ho K-shell absorption edge, in order to reduce its fluorescent contribution (it cannot be entirely eliminated due to the third-harmonic reflection of the Si monochromator). The sample was mounted in a 0.6 T field in the 10 mm air-gap of a conventional electromagnet. Previous magnetization measurements at room temperature, on a sample of almost identical shape and size, taken from the same polycrystalline plate, had shown that this field is sufficient to produce more than 80% saturation of the sample. Our data are very close to those recorded on a $HoFe_2$ crystal sample with fields up to 14 T at room temperature, where saturation in (001) direction was reached already at 0.2 T [16]. At 0.6 T the saturation for (110) and (111) directions can be estimated from the same data to 80% and 70%, respectively. Surface magnetization effects, which might be serious in low-energy diffraction studies, are insignificant in Compton scattering where the 48 keV beam penetrates several hundred microns in $HoFe_2$.

The electromagnet was constructed with a 4 mm axial hole through one of the pole pieces so that the magnetic field could be aligned with either the incident or scattered beam wave-vector. The scattering angle was chosen to be 90°. The same geometry has been used for magnetic diffraction studies on $HoTbFe_2$ sample [17] where the authors report evidence for the separation of spin and orbital contribution to the sample magnetization. For this geometry equation (1), apart from the leading terms, takes the form

$$\boldsymbol{F}_S\,[2\sin\alpha + \varepsilon(\cos\alpha + \sin\alpha)] + \boldsymbol{F}_L\,[\sin\alpha + (1 + \varepsilon)\cos\alpha)]\,. \qquad (4)$$

Inspection of equation (4) shows that in this geometry the contributions to the scattering are

$$\propto [\boldsymbol{F}_S\varepsilon + \boldsymbol{F}_L(1+\varepsilon)]$$

with the field parallel to the incident beam ($\alpha = 0°$, '$\boldsymbol{F}_L$ geometry') and

$$\propto [\boldsymbol{F}_S(2+\varepsilon) + \boldsymbol{F}_L]$$

with the field parallel to the scattered beam ($\alpha = 90°$, '$\boldsymbol{F}_S$ and $\boldsymbol{F}_L$ geometry').

In our experiment $\varepsilon = -0.086$. The two signals should be very different because in $HoFe_2$ $\boldsymbol{F}_L$ is some three times larger than $\boldsymbol{F}_S$, at room temperature, and is of the opposite sign. The orbital moment comes almost entirely from the 4f Ho electrons whereas the spin moment resides largely on the Fe sites and is antiferromagnetically coupled to the Ho moment. This difference in origin (4f rather than 3d) also leads to a characteristic difference in the two momentum distributions.

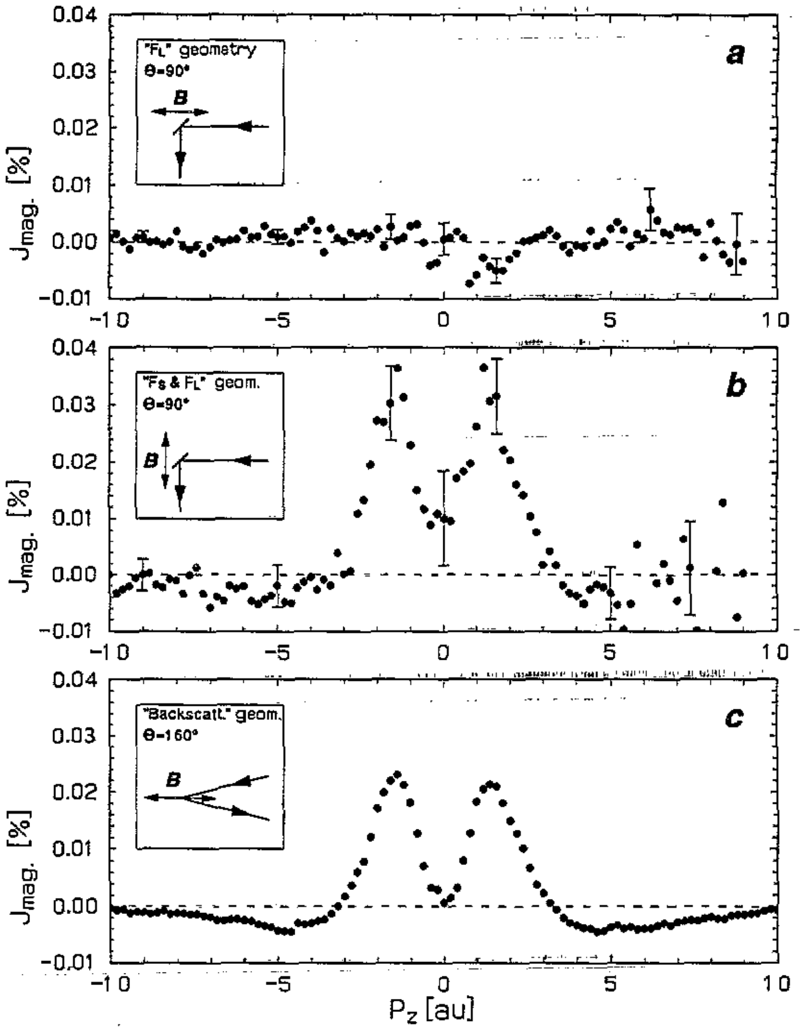

Figure 1. Magnetic Compton profile of the $HoFe_2$ sample defined as $(J^+ - J^-)/(I^+ + I^-) \times 100\%$, where $J^\pm$ are the total Compton profiles for each spin direction, and $I^\pm$ are their respective integrals from -10 au to $+5$ au (the range of integration for positive momenta is limited to $+5$ au to avoid strong fluorescent peaks in 90° scattering). Positive and negative superscripts correspond to the different directions of magnetic field. The geometrical configurations and magnetic field directions are shown in the insets. (a) $\boldsymbol{F}_L$ geometry; the expected large magnetic contribution from the orbital magnetization is not seen. (b) $\boldsymbol{F}_S$ and $\boldsymbol{F}_L$ geometry; the positive magnetic profile follows the spin-only line-shape measured in the backscattering geometry adopted in (c) below. (c) Backscattering geometry; the spin-only contribution to the magnetic Compton profile can be measured and the line-shape is the same as found in (b) above. Statistical errors are of the size of the data points.

The magnetic Compton profile is formed by taking the difference between the spectra with the field alternately parallel and antiparallel to the chosen direction which eliminates the contribution from the spin-paired electrons. The results, after conversion to an electron momentum scale, are shown in figure 1. No magnetic profile is evident above the statistical noise in the F_L geometry (upper diagram). If a 4f orbital contribution of the magnitude 3–4 μ_B [12] were present this profile would show single broad peak of similar height to that in part (*b*); furthermore there would be no central dip. In the F_S and F_L geometry (middle diagram) there is a significant signal. This is in fact a spin-only magnetic profile as is clear from two observations. Firstly, it is of the opposite sign to what would be expected if the dominant F_L contribution was present (the sign of the profile was established by a measurement on a soft iron). Secondly, it has the same line-shape as the spin-only profile which was measured in 'backscattering' geometry in a separate experiment (see lower diagram) and successfully analysed in terms of Ho and Fe spin-dependent momentum distributions; those results will be reported elsewhere [18].

This is clear evidence for the fact that in magnetic Compton scattering experiments of this type the orbital contribution is either very much reduced (by more than a factor of ten) from the value predicted in [2] or absent. These experiments are carried out, for a variety of reasons, at energy transfers that are large compared with the binding energies of the electrons responsible for the ferromagnetism. They are usually interpreted within an impulse approximation which is associated with an interaction that is 'fast' on the timescale of, say, the period for classical orbital motion and this has been taken by some as inferring that orbital magnetization will not be measurable in this type of experiment: such semiclassical arguments may, however, be misleading. Whether such a contribution would be evident at lower energies and smaller energy transfers is not clear, but it would be difficult to interpret data taken under those conditions. The present results do not rule out the use of magnetic Compton scattering as a method of separating F_L and F_S, if they can be made on an absolute scale, since the F_S can be measured and F_L then deduced from F_S and F_L data.

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