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1995 J. Phys.: Condens. Matter 7 L175

(http://iopscience.iop.org/0953-8984/7/13/002)

J. Phys.: Condens. Matter 7 (1995) L175–L180. Printed in the UK

LETTER TO THE EDITOR

Enhanced magnetic x-ray diffraction from a nearly perfect crystal of iron

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Received 24 January 1995

Abstract. We report the first non-resonant magnetic x-ray diffraction experiments to be performed at the European Synchrotron Radiation Facility, the world's first third-generation synchrotron source, where we have measured the magnetic–charge interference scattering from a single crystal of iron. Pure non-resonant magnetic x-ray scattering is so weak as to be practically unobservable in ferromagnets where charge and magnetic reflections are superimposed. However, the magnetic–charge interference scattering is predicted in kinematic theory to be a $\sim 0.1\%$ effect, and was previously measured by Collins *et al* on a mosaic crystal of iron using the Daresbury source. We have used a nearly perfect crystal of iron, and have observed an overall enhancement of the magnetic interference signal by an order of magnitude over the previous measurement. The crystal appears to be scattering dynamically, and the enhanced interference term exhibits several interesting features which we attribute to dynamic effects. The enhancement of the magnetic signal allows remarkably fast data collection; a high quality interference spectrum for the $(h, 0, 0)$ series being obtained in ~ 20 minutes counting.

The advent of high-intensity synchrotron sources has made magnetic x-ray diffraction a potentially viable alternative to neutron methods in the study of magnetic structures, and unpaired-electron distributions. Pure non-resonant magnetic x-ray scattering [1, 2] is extremely weak, a factor of $\sim 10^{-6}$ of the charge scattering, and as a result most studies undertaken so far have relied on the 'anomalous' enhancement of the magnetic scattering near to K and L shell binding energies [3]. It is, however, useful to explore the possibilities of measuring non-resonant scattering, as this is a completely general effect.

The weakness of the magnetic scattering is a particular problem in the case of ferromagnets, where magnetic and charge reflections are superimposed. However, the magnetic–charge interference term is predicted in kinematic theory [1] to be a $\sim 0.1\%$ effect, and has been measured by Collins *et al* [2] on a mosaic crystal of iron, using a field switching technique at the Daresbury source.

In this letter we report equivalent experiments on a nearly perfect crystal of iron, where we have observed an enhancement in the overall magnetic interference term by an order of magnitude for low-order reflections and by an even larger factor for high-order ones. The dependence of the effect on the polarization of the incident beam differs significantly from that predicted by the kinematic theory, and the rocking curve of the magnetic interference

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0953-8984/95/130175+06$19.50

term is significantly sharper than that of the pure charge scattering. A likely cause of these effects is dynamic scattering, resulting from the high crystalline perfection of the sample.

In kinematic theory [1], the cross section for 90° diffraction from a centrosymmetric ferromagnet, with the magnetization saturated along the diffracted beam direction, is to first order

$$d\sigma/d\Omega = \left(e^2/mc^2\right)^2 \tfrac{1}{2}\left((1 - P_l)|F_c|^2 - g P_c F_c' F_m + g P_{45} F_c'' F_m\right) \qquad (1)$$

where $g = E/mc^2$ (= energy (keV)/511), and F_c' and F_c'' refer to the real and imaginary parts of the charge form factor $F_c = F_c' + iF_c''$ [4]. F_m is the corresponding magnetic form factor. For a horizontal scattering plane the polarization of the incident x-ray beam can be represented as a sum of circular (P_c), linear in-plane (P_l), and linear 45°-inclined (P_{45}) components. In general the synchrotron radiation is almost entirely linearly polarized in the orbit plane and partly circularly polarized off-orbit, P_{45} being negligible. The signs of the interference terms in equation (1) can be reversed by reversing the applied field. Since the third term in (1) is negligible for synchrotron radiation [4], one obtains the flipping ratio or normalized difference profile R [2]

$$R = (I^+ - I^-)/(I^+ + I^-) = g(P_c/(1 - P_l))(F_m(k)/F_c(k)) \qquad (2)$$

where I^+ and I^- denote the diffracted intensity for the two field directions. From (2) we see that R is proportional to the incident photon energy and the ratio of the magnetic to charge form factors.

We have performed a total of four experiments, using the white beam provided by two different stations, D5 and ID15, at the European Synchrotron Radiation Facility [5]. The optics beamline, D5, using a 0.85 T bending magnet allowed measurement of high-quality magnetic diffraction peaks up to the (14, 0, 0) order, whilst the high-energy beamline 1.85 T asymmetric wiggler on ID15 provided increased flux for measurement to the previously unobserved (22, 0, 0) magnetic diffraction order. The bending magnet [5] provides a polarization ratio $P_c/(1 - P_l)$ of about 6.5 at 18.34 keV, corresponding to the (6, 0, 0) reflection of iron, for a 0.1 mm slit positioned 0.5 mm off-orbit 33 m from the source.

The single cubic iron crystal of very low mosaicity (< 10″) [6], was studied in Bragg scattering geometry with $2\theta = 90°$. A magnetic field sufficient to saturate the sample (±400 G), was applied parallel to the scattered beam by means of a specially designed electromagnet with hollow pole pieces. Sample movement and field-dependent strain effects were estimated to be negligible [7]. To avoid over saturation of the Ge solid state detector, positioned 1.5 m from the sample, the incident beam was slitted down to typically ~ 200 μm horizontally $\times$ 100 μm vertically, and a vertical slit variable between $\sim$ 50–400 μm was positioned at the entrance of the detector [8]. Periodic reversal of the sample magnetization every 5 s allowed the magnetic interference signal to be accurately determined from the difference spectra. The normalization and subtraction technique, which was carefully controlled, allowed magnetic peaks to be observed up to the (22, 0, 0) order. Figure 1 shows the (h, 0, 0) series of charge diffracted peaks obtained in some 600 seconds, using the ID15 asymmetric wiggler, and the corresponding multichannel analyser (MCA) difference spectrum ($I^+ - I^-$) for the magnetic scattering. Integration of the difference spectrum clearly showed that only the Bragg reflections contribute to the difference profile, the fluorescence and escape peaks integrating to zero. This confirms the absence of all systematic errors except possibly sample movement [7].

Using a very narrow slit (~ 50 μm) in front of the detector, rocking curve measurements showed very narrow reflection widths of less than $\sim$ 10″ ($\sim$ 20 times the intrinsic Darwin width), indicative of a very high quality iron crystal with a small mosaic [6]. For a mosaic

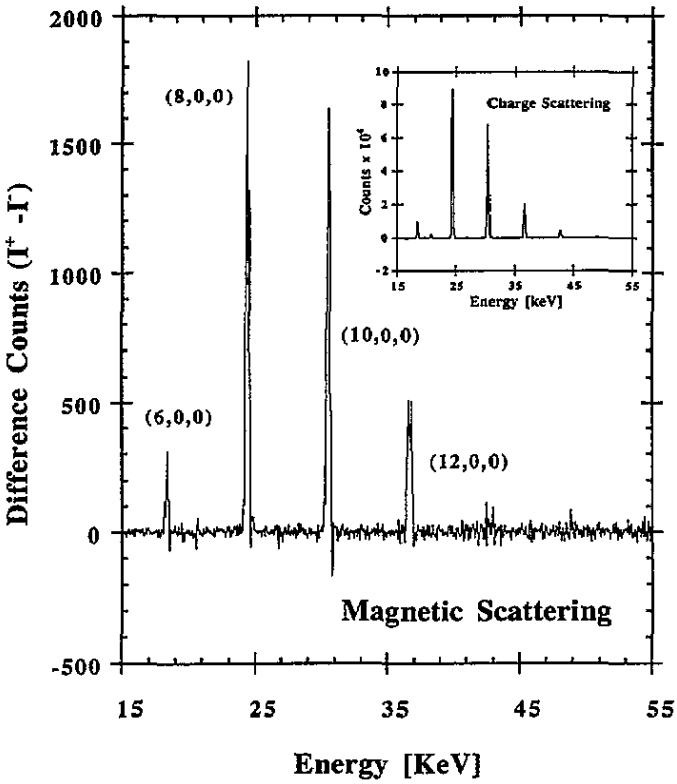

Figure 1. Typical magnetic difference profile obtained for (h, 0, 0) reflections of iron as a function of energy. Data were collected using the ID15 high energy asymmetric wiggler, with a 3 mm aluminium absorber placed within the incident beam to attenuate low-energy reflections. Inset: corresponding charge diffraction spectrum, measured with 10 minutes counting time.

crystal we would expect rocking curve widths in the order of minutes, rather than seconds of arc. The perfection of the crystal has recently been confirmed by x-ray topographic measurements [9]. In order to vary the off-plane polarization and hence the polarization ratio, the whole diffractometer was scanned vertically across the incident beam and magnetic diffraction spectra measured for different heights. A small slit $\sim 50\ \mu$m was used in front of the detector, and at each height the crystal was carefully realigned on the maximum of the rocking curve. The scan for the (600) peak at 18.34 keV using the bending magnet source is shown in figure 2, where the flipping ratio $R = (I^+ - I^-)/(I^+ + I^-)$ is displayed as a function of vertical sample position relative to the beam centre. The inset in figure 2 shows the calculated polarization ratios, the accuracy of which has been confirmed by magnetic Compton scattering, an incoherent process [10]. According to equation (2), R should scale with the ratio $P_c/(1 - P_l)$, which is antisymmetric above and below the orbit plane. The observed effect, in contrast, is clearly asymmetric, with a significant flipping ratio $R \sim 0.2\%$ at the beam centre, where $P_c = 0$. The maximum in $|R|$ of $\sim 0.8\%$, positioned ~ 0.5–1 mm off-orbit, is 8–9 times larger than would be expected from the kinematical result given in equation (2) and observed at Daresbury [2]. The interference scattering measured for the different reflection orders from (2, 0, 0) to (20, 0, 0) are compared with the kinematical prediction in figure 3. Since the polarization ratio $P_c/(1 - P_l)$ only changes slowly with energy, the ratio R/g is approximately proportional to the ratio of the magnetic to charge

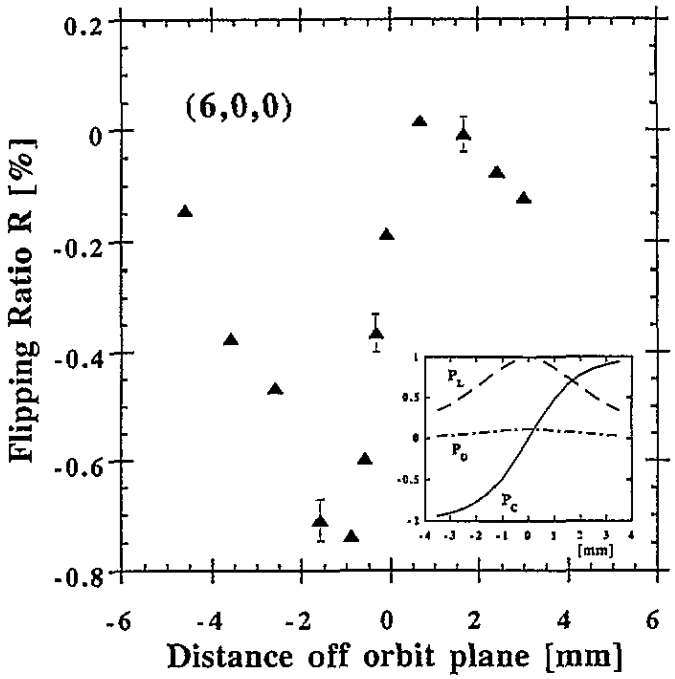

Figure 2. Observed flipping ratio $R = (I^+ - I^-)/(I^+ + I^-)$ for the (6, 0, 0) reflection as a function of distance in mm off the synchrotron orbit plane. The data were measured at a distance of 33 m from the bending magnet source, using a 0.1 mm vertical slit. Inset: The calculated degrees of polarization (linear P_l, circular P_c and unpolarized P_u) as a function of distance of orbit in mm.

form factors [1]. Qualitatively, the observed data falls off with increasing energy as in the kinematical model, but a quantitative comparison at high momentum transfer is difficult as no recorded data, experimental or theoretical, exists above the (6, 0, 0) order. It is evident, however, that for all energies the observed magnetic effect is far bigger than that expected from the kinematical model and at very high momentum transfers the effect remains very large. These results were measured at the peak of the charge diffracted beam using a very small slit of $\sim 4''$ of arc. Further investigation showed that the enhanced magnetic effect decreases as an integration is performed over the charge reflection by increasing the slit size in front of the detector. This is shown (inset, figure 3) for the (600) reflection, and clearly indicates that the magnetic diffraction width is much narrower than that of the charge reflection.

We believe that the above experimental results are probably explained by dynamical scattering within the iron crystal. The anomalous polarization dependence suggests either that there is a second, unexpected contribution to the interference term, or that the incident beam suffers a phase shift within the perfect crystal which changes the effective polarization [11]. We note that the asymmetry of the Bragg scattering geometry means that reversing the polarization is not simply equivalent to reversing the field, so there is no symmetry reason why the interference effect should be rigorously antisymmetric above and below the orbit plane.

The overall enhancement of the magnetic signal is due in part to the narrowing of the magnetic interference scattering rocking curve with respect to that of the pure charge

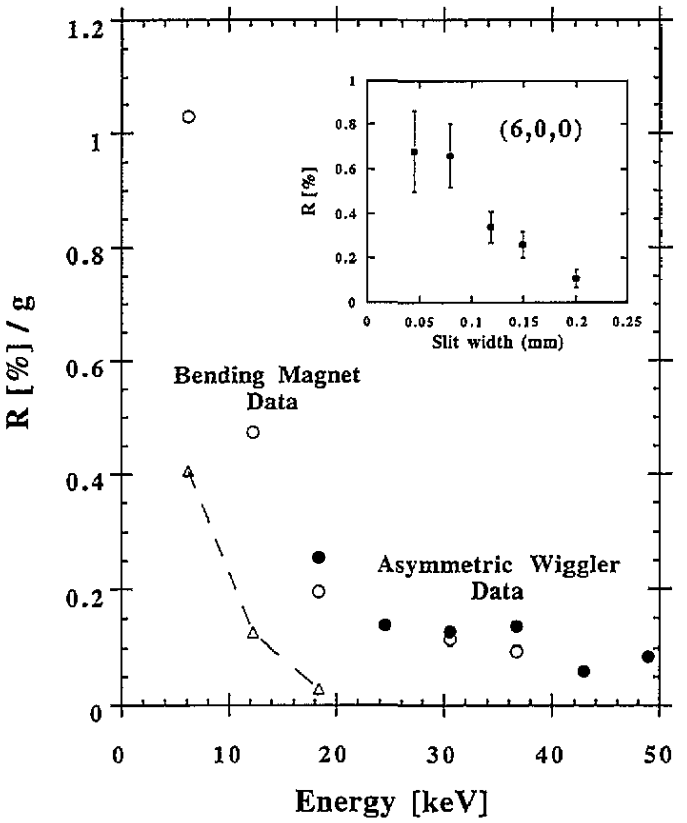

Figure 3. Observed flipping ratios divided by the incident beam energy $-R/g = -(I^+ - I^-)/(I^+ + I^-)(E/mc^2)$ as a function of energy for the (h, 0, 0) orders. Shown are the data measured using the bending magnet source (open circles) and the asymmetric wiggler (full circles). For the purposes of comparison the asymmetric wiggler data has been scaled with respect to the bending magnet results so the (800) reflections are equivalent. Also shown (triangles) are the values calculated using the kinematic theory [1]. Inset: Flipping ratio of the (6,0,0) reflection as a function of the width of the slit in front of the detector, corresponding to partial integrations over the rocking curve. Approximately 0.2 mm corresponds to an integration over the entire reflected beam. The data in the main part of the figure were measured using a 50μm slit with the sample positioned 2 mm below the orbit plane.

reflection. This suggests that the enhancement of the interference term occurs only very close to the Bragg condition, which is consistent with a purely dynamical explanation. We have not yet been able to characterize accurately the rocking curve, but initial results suggest that the true profile of the interference contribution may exhibit complex structure, again consistent with dynamical scattering.

Even when integrating entirely over the charge and magnetic reflections, the flipping ratio R is still several times ($\sim$ 3 times for the 600 reflection) that expected from kinematical theory. Primary extinction effects might contribute to this enhancement. For a 90° scattering geometry, extinction occurs for the polarization component perpendicular to the diffraction plane. It is easy to show that the polarization ratio $P_c/(1 - P_l)$ rises exponentially into the crystal, giving different effective scattering volumes for magnetic and charge scattering.

In conclusion, the enhanced magnetic interference scattering we observe for non-

resonant x-ray diffraction from a nearly perfect iron crystal is difficult to explain within the context of the kinematical theory developed in recent years [1]. Dynamical scattering effects may play an important role in modifying and enhancing the interference scattering. The large magnetic effects measured here ($\sim 1\%$ of the charge scattering) are particularly interesting because they demonstrate the possibility of accurate magnetic structure and form factor determination at new third generation synchrotron sources using non-resonant magnetic diffraction techniques. Although the magnetic effects are still smaller that those obtained by conventional neutron diffractometry techniques, the very high x-ray flux available, which is primarily limited by solid-state detector count rates, allows magnetic diffraction spectra to be collected to very high order in less than one hour. We would both like to thank the

European Synchrotron Radiation Facility for allowing access to the D5 Optics and ID15 high energy beamlines at such an early stage in the commissioning process. In particular it is a pleasure to thank Pekka Suortti, Andreas Freund, Veijo Honkimaki and Ulrich Lienert of the ESRF for their help and advice in making these measurements.

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[6] Cubic $5 \times 5 \times 5$ mm^3, faces (1, 0, 0); grown by Goodfellows Ltd (UK) to $> 99.95\%$ purity.
[7] Tests for sample movement included: (1) Rocking curve measurements for the two magnetization directions, (2) direct measurement of movement by laser reflectometry, (3) studies of restoring force on sample, (4) the measurement of zero effect at certain table heights.
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