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LETTER TO THE EDITOR

Magnetic near-edge structure in iron

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Abstract. The spin-dependent variation in the absorption of circularly polarised x-rays near the K edge in ferromagnetic iron has been measured at the Daresbury storage ring by monitoring the fluorescence signal. The magnetic modulation of the absorption coefficient is found to be approximately twice as large as predicted by a first-principles spin-polarised band calculation. This discrepancy is now known to haye been present in earlier transmission data.

The measurement of x-ray absorption near-edge structure (XANES) is an established method of probing the local density of states immediately above the Fermi energy (see, for example, Brown and Doniach 1980), the dipole selection rule dictating the nature of the states that can be studied. In ferromagnets there is a possibility of differentiating between the majority and minority bands if the photo-electron can be spin polarised. This occurs with the absorption of right- and left-hand circularly polarised radiation which partially polarises the photo-electron along the direction of the photon beam. Therefore, in an experiment in which the photon beam is parallel/antiparallel to the magnetisation direction, the transition probabilities to the majority and minority bands differ.

The theory of magnetic photo-absorption in iron has been presented by Ebert *et al* (1988) in an approach that treats relativistic and spin-polarisation effects on an equal basis and arrives at an expression for the spin-dependent absorption coefficient $(\Delta\mu)_m$.

The calculations of Ebert *et al* (1988) were prompted by the first K-edge absorption measurements with circular polarisation, which were made on iron by Schutz *et al* (1987) at the DESY synchrotron. Elliptically polarised beams 0.13 mrad above and below the orbital plane were selected by slits and monochromated by a 311 channel-cut Si crystal to provide simultaneous absorption measurements of both left- and right-handed polarisations through an iron foil, 2.5 μm thick, magnetised in its plane at an angle of 30° to the incident beam direction. The magnetisation direction was reversed frequently to counteract the intensity variations associated with the decay of the beam—data for 'up' and 'down' spin alignments being stored in separate memories. The same approach has recently been applied to the study of rare-earth ferromagnet L edges where the magnetic modulation of the transmitted intensity is generally one to two orders of magnitude higher (10^{-2} as against 10^{-4})—see Schutz *et al* (1988).

The measurements reported here and performed at the Daresbury storage ring used a similar 'inclined view' method, developed for magnetic Compton scattering studies (Cooper *et al* 1988), to extract the circularly polarised flux. The apparatus was some 80 m distant from the tangent point of a bending magnet producing synchrotron radiation with a critical energy of 3.2 keV. The optimum magnetic signal was obtained by selecting a beam with a minimum inclination of 0.125 mrad to the orbital plane prior to monochromatisation by a vertically dispersing Si 111 channel-cut crystal. The synchrotron flux at 21 keV, which is seven times the critical emission energy of the bending magnet, is vanishingly small. Therefore there is no parasitic fluorescence contribution excited by the Si 333 reflection, which might otherwise distort the measured intensity ratios. Making simultaneous measurements above and below the plane was not possible, but the good positional stability of the beam in routine operation of this storage ring made that unnecessary. Data taken over several days at points where the magnetic modulation was a rapidly varying function of photon energy were found to be reproducible within their statistical accuracy. Similarly repeated scans through the 7.111 keV Fe absorption edge confirmed that the energy calibration remained fixed within the energy resolution of the slit and monochromator arrangement (approximately 1 eV at the K edge). The frequent reversal of the magnetic field not only isolates the spin-dependent signal but also eliminates many of the systematic errors associated with conventional absorption measurements (the experimental technique is similar to that described in the work of Cooper *et al* (1988) and in references therein).

This 'absorption' experiment differed from the earlier one at DESY in using fluorescence monitoring with a solid state Ge detector at 90° to the incident beam rather than transmission measurements with ionisation chambers before and after the sample: the signal, therefore, arises entirely from K-shell processes. The iron foil, again oriented at 30° to the incident beam, was only 1 μm thick. The larger magnetic modulation of the signal in the fluorescence mode, due mainly to the higher degree of circular polarisation of the beam (see below), is shown in figure 1 where both unprocessed data sets are reproduced. The statistical accuracy in the scattering experiment was necessarily inferior (the detected flux was smaller than the transmitted flux) and there was no opportunity to increase the signal level in the course of this particular measurement by using a detector with a large solid angle or a high-count-rate capability.

The other difference arose from the choice of monochromator, Si 111 in this experiment rather than Si 311 in the other. The smaller Bragg angle is important when vertical dispersion is used, since P_c is diminished upon reflection (conversely horizontal dispersion, if it were practicable, could be used to enhance the circular polarisation). In this trial experiment there was no opportunity to make an independent measurement of P_c but previous experience with magnetic Compton scattering studies have confirmed that the polarisation state of the inclined beam is well described by calculations made by one of us (Laundy 1988) based on the high-brightness magnet lattice emittance. An initial value of $P_c = 0.6 \pm 0.1$ becomes $P_c = 0.5 \pm 0.1$ after the 111 reflections whereas $P_c = 0.9 \pm 0.1$, quoted by Schutz and co-workers, should be 0.25 after two 311 reflections. This last value is a correction to that quoted in their paper, 0.42 ± 0.06, which is now known to be in error (Schutz 1988).

The absorption rates have been rescaled accordingly and plotted in figure 2 for comparison with the Daresbury data. The origin of the energy scale for the latter was chosen to be consistent with that for the DESY data. The experimental results are in satisfactory mutual agreement, bearing in mind the fact that the absolute scale depends on a knowledge of P_c and in both cases this was uncertain to 10–20%. Now, however, there is a significant difference between the experiments and theory that was not evident

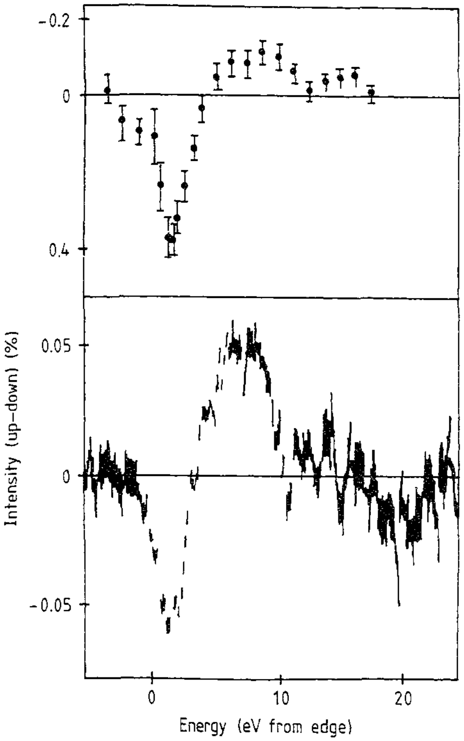

Figure 1. The upper panel shows the magnetic modulation of the fluorescence scattering in the near-edge region as directly measured in this experiment as a function of the photo-electron energy. The data points below the edge correspond to the filling of empty states through the resonant Raman process (see Manninen *et al* 1986). Above the edge the data collection time was approximately 30 minutes per point. The lower panel shows the intensity variations recorded in the transmission experiment at DESY (Schutz *et al* 1987). Note that the magnetic modulations are of opposite senses in the complementary fluorescence and transmission experiments—hence the scale inversion.

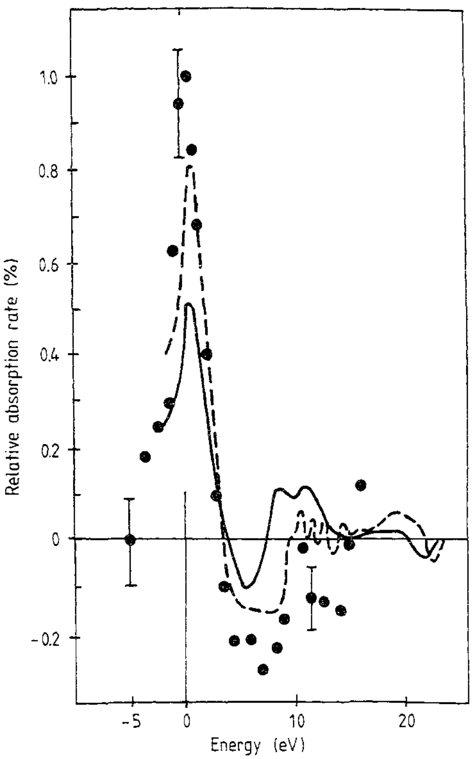

Figure 2. The relative magnetic absorption rate $(\Delta\mu)_m/\mu$ is plotted as a function of photo-electron energy; the zero corresponds to the Fermi energy. The full circles represent the fluorescence data obtained at the Daresbury storage ring and the broken curve is the DESY result (Schutz *et al* 1987) rescaled as described in the text. The full curve is from the first-principles spin-polarised band calculation of Ebert *et al* (1988).

at the time when the latter was published; it concerns the scale of the effect. The peak immediately above the Fermi surface is closer to a 1% relative absorption rate than $\frac{1}{2}$%, confirming that the calculation underestimates the magnetic absorption rate. Furthermore, neither data set shows any evidence for the second positive peak at around 10 eV. (Note that the labels on the experimental and theoretical curves in figure 1(*b*) of the paper by Ebert *et al* (1988) were inadvertently transposed.)

In summary, these fluorescence results confirm the published transmission measurements, after the latter have been rescaled to incorporate a new polarisation correction; furthermore, recent, more precise transmission data (Schutz 1988) are consistent with these results. The agreement with theory is now not as good as would have been supposed on the basis of the earlier data set although, within the context of a first-principles calculation, it is as good as might be expected. The results demonstrate the feasibility of spin-polarised absorption measurements with circularly polarised radiation extracted by the inclined view method, especially when the storage ring has good orbital stability. The fluorescence method is best suited to dilute magnetic systems and could undergo many refinements that were out of the question in this trial experiment. The extension of measurements to the higher-energy EXAFS range would also allow the local magnetic environment in alloys to be probed. Studies that could take advantage of the much higher magnetic cross sections associated with L edges would be particularly effective.

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