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A Technique for Measuring the Magnetic Disorder Scattering of Neutrons

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There is interest in measuring the contribution of magnetic origin to the background scattering of neutrons by magnetic alloys. A discussion is presented of the proper choice of experimental conditions for work with short-wavelength neutrons and of the corrections which must be applied to the data. Practical results are quoted, confirming the analysis and illustrating the procedure used in studies of Fe-Ni reported in a companion paper.

1. A neutron scattering technique of considerable current interest in magnetism is that in which a weak component of elastic magnetic scattering is separated out from a variety of other contributions to the total diffraction pattern of a magnetic substance. This technique, first exploited by Shull and Wilkinson[1] in 1955 and now by several authors at the present Conference[2)3)], makes use of the fact that the elastic magnetic disorder scattering cross-section of a spin system can be switched off by so rotating the spins that they lie along the scattering vector for which the counter is set at a given moment. In the absence of complications, the required intensity distribution is easily measured as the difference between the respective diffraction patterns with the specimen magnetised along and, say, perpendicular to the scattering vector.

The resulting difference-distribution may show Bragg peaks, and will in general show a continuous distribution of background intensity whose angular variation may or may not be indicative of local order in the spin system. In the complete absence of local order, such a background is isotropic except for the angular variation imposed by the Debye-Waller factor and by the form factors f of the atoms contributing. A case of the latter kind is FeCr[1]. If the two types of atom be assumed to have characteristic magnetic moments μ_A and μ_B, and if c is the concentration of one of the constituents, then the magnetic elastic disordered scattering (meds) differential cross-section in such a case is

$$\frac{\partial\sigma}{\partial\Omega}=0\cdot073q^2\cdot c(1-c)(\mu_A f_A-\mu_B f_B)^2 \qquad (1)$$

barn. As usual, $q^2=1-(\boldsymbol{e}\cdot\boldsymbol{\kappa})^2$ with $\boldsymbol{e}$ the unit scattering vector and $\boldsymbol{\kappa}$ a unit vector in the direction of magnetisation. Shull and Wilkinson have used measurements of this quantity to determine $(\mu_A-\mu_B)^2$ for Fe Cr and other alloys, thereby throwing light on the values of μ_A and μ_B separately.

If the magnetic sample be a single crystal of such dimensions and shape that multiple scattering is unimportant, and if the neutron wavelength be such as to minimise inelastic scattering, then it may be that there are no further complications and the above formula can be applied to the measurements directly. More often, however, to obtain a quick result a sample is used that is not a single crystal, and if the specimen contains a fair number of grains there will be a danger that Bragg scattering, or, worse, multiple Bragg scattering will take place and cast doubt on the measurements. In such a case the only practicable resort is to use a preparation of such small grain size that it is a perfect polycrystal. Debye-Scherrer powder-diffraction will then occur, and multiple scattering will distort the difference-pattern obtained by subtracting the patterns taken under two different conditions of magnetisation. The corrections for the extra effects can be as large as the meds effect itself, but they can be applied systematically.

A discussion of the corrections necessary when short-wavelength neutrons are used is given in what follows.

2. In a typical experiment the cross-section sought will be of the order of 0.01 barns sterad^{-1} atom^{-1}, whereas the Debye-Scherrer cross-section will be of the order of 0.4.

There may also be a nuclear elastic disorder cross-section of the general order 0.05. The desired intensity, therefore, will be in competition with large quantities of other types of scattering. When the magnetisation direction is switched, to obtain a difference count, two types of undesirable effect will take place: (i) due to the single transmission effect[4], the overall exposure of the body of the specimen will be altered; and (ii) a contribution to the difference count will be made by neutrons magnetically scattered at wrong angles but deflected into the counter by further scatterings. It will be argued that effect (i) is very small in a practical case, where effect (ii) has been properly allowed for.

All the forms of multiple scattering must be considered in an actual experiment. To illustrate the arguments involved, consideration will be given to two: multiple Bragg scattering and multiple disorder scattering.

Multiple Bragg scattering

If we represent the single Bragg scattering as giving rise to a set of cones radiating from some point as origin, the directions of double Bragg scattering are the directions obtained by setting up this family of cones once more around every ray of the first set. If a process $\tau \equiv hkl$ is followed by another, $\tau' \equiv h'k'l'$, both for simplicity say of small Bragg angle, the distribution of intensity recorded on an imaginary photographic plate in the forward direction would be as shown schematically in Fig. 1. A counter

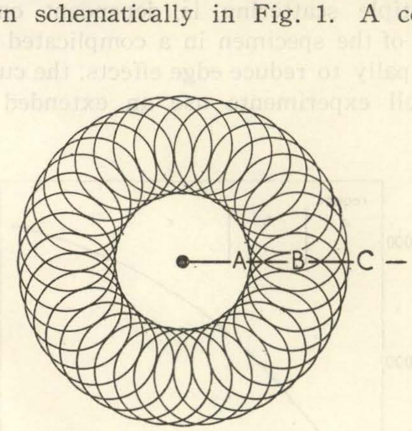

Fig. 1. Schematic representation of the family of Debye-Scherrer cones created by a double Bragg scattering process in a polycrystal, and projected onto a photographic plate in a neutron diffraction experiment. The main beam strikes the plate in the centre.

tracking along a direction in the equatorial plane represented by a horizontal line in this figure would pick up the $\tau\tau'$ process as a band of intensity between the two angles $|\theta_B \pm\theta_B'|$; at the two extremes of the band there would be a singularity in the intensity.

Fig. 2 shows the observed diffraction pattern of a plate of polycrystalline iron, 3 mm in thickness, taken in connexion with some studies on the Fe-Ni system[3]. A spectrum of lines at the top of the figure shows the position of the expected singularities due to

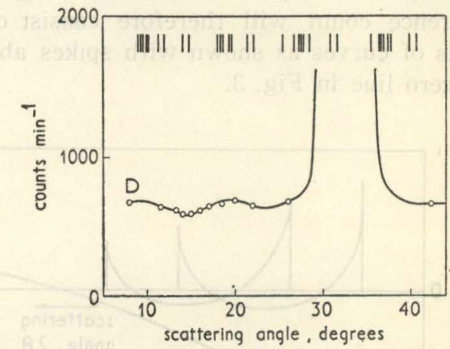

Fig. 2. The diffraction pattern of a specimen of polycrystalline iron, $1.59 \times 1.59 \times 0.30$ cm^3 in dimension, corrected for background and nuclear disorder scattering. In addition to the 110 reflexion there is apparent a highly irregular level of diffuse intensity, almost all due to double Bragg scattering. Calculated positions where, with perfect resolution, peaks would be found in this diffuse intensity are shown by a band of lines in the upper part of the figure.

various $\tau\tau'$ combinations, and it will be seen that even with an instrumental width designed (for a reason given later) to broaden the single-Bragg peaks to a base-width of 7°, there is still a very marked undulation in the background. In the Fe-Ni experiment, the interesting meds was normally about 2% of a background intensity of this general magnitude. Inelastic scattering is also present, but may be ignored for the purposes of this paper.

It will be evident from Fig. 1 that at A and C at the extremes of a $\tau\tau'$ band, both Bragg scatterings are in the equatorial plane. Since the angular range of scattering over which magnetic processes are considerable is only a few tens of degrees, the scattering vectors $\boldsymbol{e}_B$ of these Bragg processes will normally be close to the $\boldsymbol{e}$ of the meds at any given setting; and therefore when the conditions $\boldsymbol{e} \parallel \boldsymbol{\kappa}$ and $\boldsymbol{e} \perp \boldsymbol{\kappa}$ are set up the magnetic part of the Bragg intensity tends to behave in the same way as the meds itself. The upshot will be a *positive* spurious effect. On the other hand, at B in the centre of the band of intensity, one of the Bragg scatterings has a more or less vertical $\boldsymbol{e}_B$. In this case the condition $\boldsymbol{e} \parallel \boldsymbol{\kappa}$ produces very nearly $\boldsymbol{e}_B \perp \boldsymbol{\kappa}$, and with vertical magnetisation $\boldsymbol{e} \perp \boldsymbol{\kappa}$ produces very nearly $\boldsymbol{e}_B \parallel \boldsymbol{\kappa}$; the result is a *negative* spurious effect. The spurious contribution of $\tau\tau'$ processes to the magnetic difference count will therefore consist of a series of curves as shown with spikes above the zero line in Fig. 3.

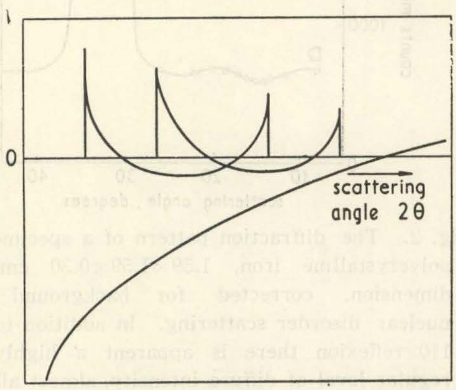

Fig. 3. Spurious contribution of various double Bragg scattering processes to the apparent magnetic disorder scattering (schematic). Plotted vertically is the difference intensity between diffraction patterns taken with the specimen magnetised perpendicular and parallel to the scattering vector.

Processes $\tau\tau$, in which the two successive Bragg scatterings employ the same Debye-Scherrer cone, make a special contribution. The $\tau\tau$ process is usually of dominant importance, partly because it includes the process in which both scatterings involve the strongest magnetic line, and also because it has a pronounced intensity singularity in the forward direction and therefore is serious at the very region of low angles where the meds is of greatest interest. Consideration shows that at low angles in the diffraction pattern, $\tau\tau$ processes give a pronounced *negative* effect, shown by the strongly negative-going line in Fig. 3. To calculate it, let there be I_0 neutrons sec^{-1} in the incident beam, and let $\varepsilon_1 I_0$ have a first Bragg scattering into some given cone. Without introducing the effect of specimen shape explicitly, say that a fraction ε_2 of these neutrons have a second scattering with the same Bragg angle θ_B. Then if the observed overall scattering angle be written 2θ, the amount of double Bragg scattering from this process in a solid angle $d\Omega$ at 2θ is $I(2\theta)d\Omega$, where

$$I(2\theta)=2^{-5/2}\pi^{-2}\varepsilon_1\varepsilon_2 I_0 \operatorname{cosec}\theta\cdot(\cos 2\theta-\cos 4\theta_B)^{-1/2}. \qquad (2)$$

Either or both of the scatterings 1 and 2 may be magnetic; and if $\boldsymbol{\kappa}$ is along the external bisector of 2θ, so that $\boldsymbol{e} \parallel \boldsymbol{\kappa}$,

$$q_1^2=q_2^2=1-\tfrac{1}{4}\sin^2\theta \operatorname{cosec}^2\theta_B . \qquad (3)$$

We thus have the situation that double Bragg scattering produces a general background to the diffraction pattern, typically two orders of magnitude larger than the effect sought, and with a large number of infinite singularities; its contribution to the magnetic difference-count fluctuates violently, with infinite contributions at the singularities and oscillating between positive and negative values elsewhere. One requirement of any practical technique of dealing with this problem must therefore be a sufficiently relaxed counter collimation to smooth out the more abrupt variations. At the present day this requirement is in harmony with the need to gain sufficient intensity to do the experiment at all, which also calls for relaxed collimation.

Multiple scattering is dependent on the shape of the specimen in a complicated way. Principally to reduce edge effects, the current Harwell experiments use an extended thin

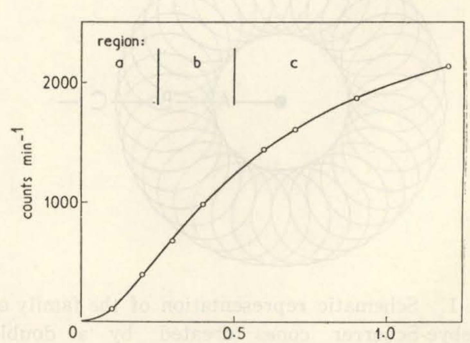

Fig. 4. The multiple Bragg scattering at 8° from a polycrystalline specimen of iron as a function of thickness.

plate in transmission; the width of the plate ($\frac{5}{8}''$) assists in creating poor angular resolution.

The intensity at the point D in Fig. 2 depends on the thickness t of the plate as shown in Fig. 4. For small t the curve is parabolic; but beyond the region a the curve falls away in region b as triple scattering becomes more frequent and double scattering correspondingly less frequent. Finally in region c the intensity tends towards saturation.

The third and higher orders of multiple Bragg scattering would, of course, need a more complicated discussion than that above, but their very complexity leads to a simplification, as the final intensities average out the oscillating effects to a great extent. Their importance in magnetic difference counting diminishes sharply with the multiplicity of the scattering. Thus, with increasing specimen thickness the spurious effect in the difference pattern must pass through a maximum value and then decrease. In connexion with the Fe-Ni experiment, this feature has been measured at the setting D with pure iron, which of course gives no meds. The result, reduced to an apparent spurious meds cross-section per atom, is reproduced in Fig. 5. Region a $(0<t<2\frac{1}{2}\text{ mm})$ is again characterised by simple behaviour, the correction being linear in t.

It is now apparent how to choose the specimen thickness. A t so small that the correction is negligible is usually inadmissible on intensity grounds; a t so great that the correction is negligible is ruled out because

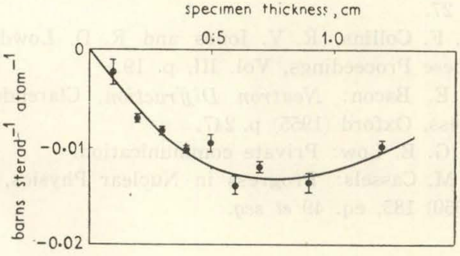

Fig. 5. A spurious contribution from multiple Bragg scattering effects to the quantity measured in an experiment to determine the magnetic elastic disorder cross-section, shown as a function of specimen thickness. The observation was taken at $2\theta=8°$ with pure iron, which has no magnetic disorder cross-section.

the desired phenomenon would be massively attenuated and dispersed in angle. The best choice is a dimension in region a where the spurious effect for an alloy has a calculable behaviour and can be corrected out by virtue of measurements taken with a pure element. For the Fe-Ni experiment of reference 3 a thickness of 2.5 mm was used, based on the data of Fig. 5; for nickel-rich alloys using Ni^{60} it was possible to increase this figure to 4 mm.

The angular distribution of the spurious cross-section was measured as a function of angle with pure iron, for which it is more serious than anywhere else in the Fe-Ni constitution diagram. Fig. 6 shows that the phenomena indicated in Fig. 3 are clearly apparent. The correction is largest at small

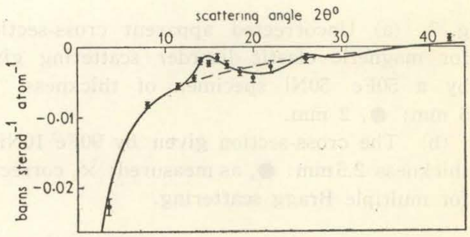

Fig. 6. The spurious cross-section of iron, as in figure 5, shown as a function of scattering angle for a plate of thickness 3 mm. A heavy line gives the theoretical shape.

angles, where it is dominated by the contribution from $\tau\tau$ processes; at higher angles the fluctuating effects, when observed with poor resolution, tend strongly to cancel out.

Having taken these measurements at one end of the constitution diagram, it remains to multiply the correction by some appropriate scaling factor so that it can be applied to alloys of general composition. For instance, if the mean nuclear scattering length $\bar{b}_{\text{nuc}}\gg p$, the magnetic amplitude, the important contributing processes will be those in which one scattering is nuclear, the other magnetic. Writing M for the intensity of magnetisation, the scaling factor would then be $(\bar{b}^2M^2)_{\text{alloy}}/(\bar{b}^2M^2)_{\text{element}}$. If the alloy has a crystal structure different from that of the element, the ε, θ_B, f and q values appropriate to any θ will also be different and a more complex scale factor must be set up, referring back to the intensity formulae. As an example, the prediction for a 50 Ni 50 Fe specimen was confirmed by measuring the difference pattern at two different specimen thicknesses, 5 mm and 2 mm, so that an empirical extrapolation to zero thickness could be made. The uncorrected apparent meds cross-sections are seen in Fig. 7a, which shows convincingly that in this (rather favourable) case the spurious effects have been made small and can be allowed for accurately.

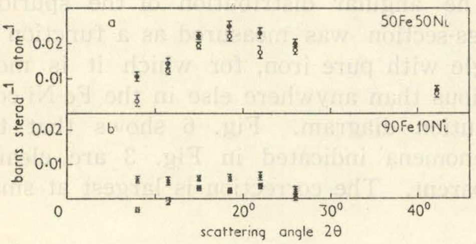

Fig. 7. (a) Uncorrected apparent cross-sections for magnetic elastic disorder scattering given by a 50Fe 50Ni specimen of thickness: ○, 5 mm; ●, 2 mm.
(b) The cross-section given by 90Fe 10Ni at thickness 2.5 mm: ●, as measured; ×, corrected for multiple Bragg scattering.

A case where the correction is more striking is illustrated in Fig. 7 b, where the raw data for a 90 Fe 10 Ni alloy are shown by filled circles. In this example the uncorrected data show almost no effect, but after correction it appears from the crosses that a significant amount of meds is in fact present.

Multiple disorder scattering

In many alloy systems there will be a large cross-section for nuclear elastic disorder scattering, either because of intrinsic spin-incoherence or because the scattering lengths of the alloy components are markedly different. Such systems will be subject to double scatterings in which one event is due to nuclear disorder, one to magnetic disorder. As disorder scattering is continuous in angle, the correction will be a smooth function of 2θ, and by virtue of the averaging over all scattering angles the response of the intensity to magnetisation of the specimen will not show the striking effects given by Bragg scattering. According to Low[5] the corrections due to multiple disorder scattering are an order of magnitude less than those due to the multiple Bragg phenomenon.

The single transmission effect

Taking the symbols from, e.g. reference 4, the single transmission effect is $\Delta I/I$, and the change in exposure of the specimen on magnetisation is $\sim\frac{1}{2}\Delta I/I$. In the region a of Fig. 4, this is small compared with the double Bragg scattering; for it is

$$\tfrac{1}{4}N^2t^2\sigma_p{}^2.$$

Now

$$\sigma_p\sim\tfrac{1}{2}N\lambda^2\sum_\tau bp(1+\sin^2\theta_B)\tau^{-1}<N\lambda^2\sum_\tau bp\tau^{-1},$$

and typically for 1 Å neutrons this would come to $\sim 0.03(p/b)N\lambda^2\sum_\tau b^2\tau^{-1}$. By a well known theorem[6], at short wavelengths

$$\tfrac{1}{2}N\lambda^2\sum_\tau b^2\tau^{-1}\sim S_{\text{nuc}}\,,$$

the ordered nuclear elastic cross-section per atom. The change in exposure is therefore

$$\approx 10^{-3}(p/b)^2N^2S_{\text{nuc}}^2t^2\,.$$

But in region a, assuming to simplify the argument that $b\ll p$,

$$NS_{\text{nuc}}t<\tfrac{1}{4}\,.$$

In a typical case, therefore, the problem is negligible.

References

1 C. G. Shull and M. K. Wilkinson: Phys. Rev. **97** (1955) 304.
2 E. O. Wollan *et al*: These Proceedings, Vol. III, p. 27.
3 M. F. Collins, R. V. Jones and R. D. Lowde: These Proceedings, Vol. III, p. 19.
4 G. E. Bacon: *Neutron Diffraction*, Clarendon Press, Oxford (1955) p. 247.
5 G. G. E. Low: Private communication.
6 J. M. Cassels: Progress in Nuclear Physics, **1** (1950) 185, eq. 49 *et seq*.