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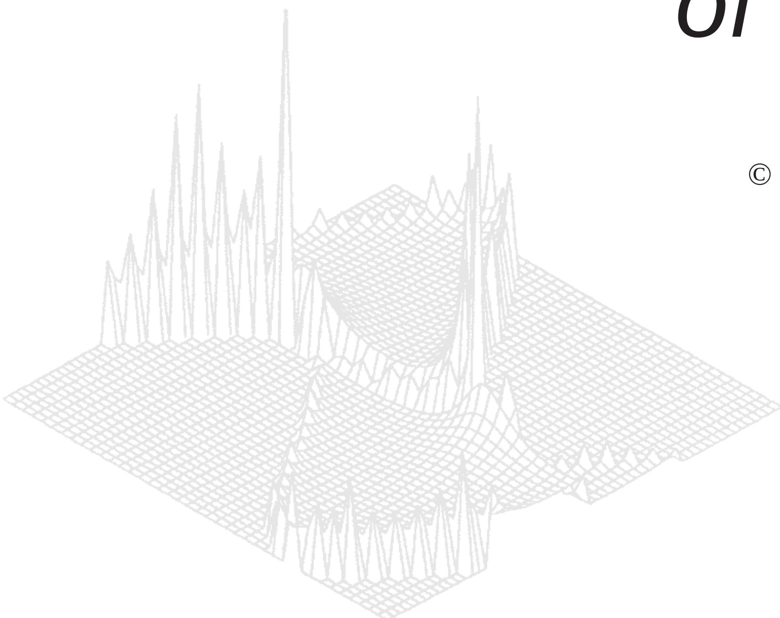

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Magnetism and Neutron Scattering*

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Abstract

Magnetic neutron scattering is very closely connected to the fundamental magnetic quantities of magnetisation and susceptibility. This is because, to a very high degree of approximation which becomes exact at long neutron wavelengths, the interaction of the neutron magnetic dipole moment is with the local magnetic induction in the magnetic material. The strength of neutron scattering for the investigation of magnetic materials lies in this and the facility of the technique to probe Fourier components of the moment and the susceptibility with wavelengths down to atomic distances, as well as to probe susceptibility fluctuation time scales the same as those that are thermally excited. This paper provides a concise account of the formal connection between neutron scattering cross sections and more familiar bulk magnetic parameters.

1. Introduction

The magnetic properties of materials are rich and varied and no one technique is sufficient to probe the whole range of behaviour. Nevertheless, neutron scattering comes closest to universal applicability. Spatial correlations are an important aspect of magnetic materials. For some techniques spatial information can only be inferred indirectly from the data, for instance by assuming that the largest effects are most probably from first neighbour atoms. Spatial information is built directly into neutron scattering experiments because of the neutron wavelength. The accessible length scale is from atomic dimensions to distances of hundreds of Ångstroms, a very useful range for correlations which involve single atoms or clusters of atoms.

The direct sensing of the magnetic induction is the other great advantage of neutron scattering for magnetic materials. There are other techniques, some of which may in particular circumstances be more sensitive, but for which there is a long chain of modelling of the interaction before a magnetic property, such as a magnetic moment value, can be inferred. One of the purposes of this paper will be to draw out the direct relationship between the neutron cross section and the magnetisation and susceptibility of magnetic materials.

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2. Interactions

The interaction of the neutron with the nucleus can be parametrised by the potential

$$\hat{V}(\boldsymbol{r}) = \left(\frac{2\pi\hbar^2}{m}\right)(b + B\hat{\boldsymbol{I}}\,.\,\hat{\boldsymbol{\sigma}})\,\delta(\boldsymbol{r}-\boldsymbol{R})\,, \tag{1}$$

where the nucleus is located at $\boldsymbol{R}$. The delta function reflects the extremely small size of the nucleus with respect to the wavelengths of thermal neutrons.

The interaction of the neutron with the magnetisation of magnetic materials is via the magnetic dipole moment of the neutron. The magnetic moment operator of the neutron can be written

$$\hat{\boldsymbol{\mu}} = \gamma\mu_{\mathrm{N}}\,\hat{\boldsymbol{\sigma}}\,, \tag{2}$$

where $\hat{\boldsymbol{\sigma}}$ is the Pauli spin matrix for spin $\frac{1}{2}$ particles, μ_{N} is the nuclear magneton, and $\gamma = -1{\cdot}91$ is the gyromagnetic ratio for the neutron. The interaction can, within a long wavelength approximation, be written in terms of the local magnetic induction $\mathbf{B}(\boldsymbol{r})$ and is given by

$$-\hat{\boldsymbol{\mu}}\,.\,\mathbf{B}(\boldsymbol{r}) = -\gamma\mu_{\mathrm{N}}\,\hat{\boldsymbol{\sigma}}\,.\,\mathbf{B}(\boldsymbol{r})\,. \tag{3}$$

This approximation, in which the sum of all the magnetic dipoles in the material is represented by the magnetic induction $\mathbf{B}(\boldsymbol{r})$, is useful when the variations being probed have a scale much larger than the individual dipoles. In cases where there is considerable orbital moment and the scale is intra-atomic (i.e. smaller than the atomic wavefunctions) the interaction with the spin and orbit of individual electrons must be considered (Lovesey 1984). However, it is in this approximation, which in many situations is very accurate, that very powerful comparisons of neutron scattering cross sections with bulk magnetic quantities like magnetisation and susceptibility can be made.

The total interaction of the neutron at a point $\boldsymbol{r}$ in a target is

$$\hat{V}(\boldsymbol{r}) = \left(\frac{2\pi\hbar^2}{m}\right)\left[b\delta(\boldsymbol{r}-\boldsymbol{R}) + \left\{B\hat{\boldsymbol{I}}\delta(\boldsymbol{r}-\boldsymbol{R}) - \frac{1}{4\pi}\left(\frac{e\gamma}{\hbar c}\right)\hat{\mathbf{B}}(\boldsymbol{r})\right\}\,.\,\hat{\boldsymbol{\sigma}}\right], \tag{4}$$

where the terms have been grouped according to the interaction with the neutron spin, and smaller terms such as those involving the velocity of the neutron and the electric field gradient have been ignored (Lovesey 1984).

In the first Born approximation the neutron cross section is given by

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega\,\mathrm{d}E}\right)_{k,k'} = \frac{k'}{k}\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{q'}\sum_{q} p_q\left|\langle\boldsymbol{k}'s'q'|\hat{V}(\boldsymbol{r})|\boldsymbol{k}sq\rangle\right|^2\delta(\hbar\omega - E_{q'} + E_q)\,, \tag{5}$$

where the neutron goes from spatial plane wave state $\boldsymbol{k}$ to plane wave state $\boldsymbol{k}'$, and from spin state s to s'. At the same time the specimen goes from state q to state q'.

We firstly consider the transition between neutron spatial states. The matrix element may be written

$$\langle s'q'| \int \mathrm{d}\boldsymbol{r}\left(\frac{2\pi\hbar^2}{m}\right)\left\{\hat{b}\delta(\boldsymbol{r}-\boldsymbol{R}) + B\hat{\boldsymbol{\sigma}}\,.\,\hat{\boldsymbol{I}}\delta(\boldsymbol{r}-\boldsymbol{R}) - \frac{1}{4\pi}\left(\frac{e\gamma}{\hbar c}\right)\hat{\boldsymbol{\sigma}}\,.\,\hat{\mathbf{B}}(\boldsymbol{r})\right\}\mathrm{e}^{\mathrm{i}(\boldsymbol{k}-\boldsymbol{k}')\,\boldsymbol{\cdot}\,\boldsymbol{r}}|sq\rangle\,, \tag{6}$$

and after inserting the plane wave states for $\boldsymbol{k}$ and $\boldsymbol{k}'$ becomes

$$\left(\frac{2\pi\hbar^2}{m}\right)\left\{\sum_{\boldsymbol{R}}[\hat{b}(\boldsymbol{R}) + B(\boldsymbol{R})\hat{I}(\boldsymbol{R})\,.\,\hat{\boldsymbol{\sigma}}]\mathrm{e}^{\mathrm{i}\boldsymbol{\kappa}\,\boldsymbol{\cdot}\,\boldsymbol{R}} - \frac{V}{4\pi}\left(\frac{e\gamma}{\hbar c}\right)\hat{\boldsymbol{\sigma}}\,.\,\hat{\mathbf{B}}(\boldsymbol{\kappa})\right\}, \tag{7}$$

where $\boldsymbol{\kappa} = \boldsymbol{k}' - \boldsymbol{k}$, and

$$\hat{\mathbf{B}}(\boldsymbol{\kappa}) = \frac{1}{V}\int \mathrm{d}\boldsymbol{r}\;\hat{\mathbf{B}}(\boldsymbol{r})\,\mathrm{e}^{-\mathrm{i}\boldsymbol{\kappa}\,\boldsymbol{\cdot}\,\boldsymbol{r}}\,, \tag{8}$$

with V the specimen volume. In the absence of an external field $\mathbf{B}(\boldsymbol{\kappa})$ is

$$\hat{\mathbf{B}}(\boldsymbol{\kappa}) = 4\pi\hat{\mathbf{M}}_{\perp}(\boldsymbol{\kappa})\,, \tag{9}$$

where $\mathbf{M}(\boldsymbol{\kappa})$ is the Fourier transform of the magnetisation distribution. The components of $\mathbf{M}(\boldsymbol{\kappa})$ parallel to $\boldsymbol{\kappa}$ are not reflected in $\mathbf{B}(\boldsymbol{\kappa})$ because of the requirement that $\nabla\,.\,\mathbf{B} = 0$. This is illustrated in Fig. 1 where the effect on $\mathbf{B}$ of magnetic dipoles alternating in sign along the moment direction is contrasted with the effect when the sign change is transverse to the moment direction. This can be used very effectively to define the direction of magnetisation in magnetic materials, such as antiferromagnets, with no gross external magnetic effects.

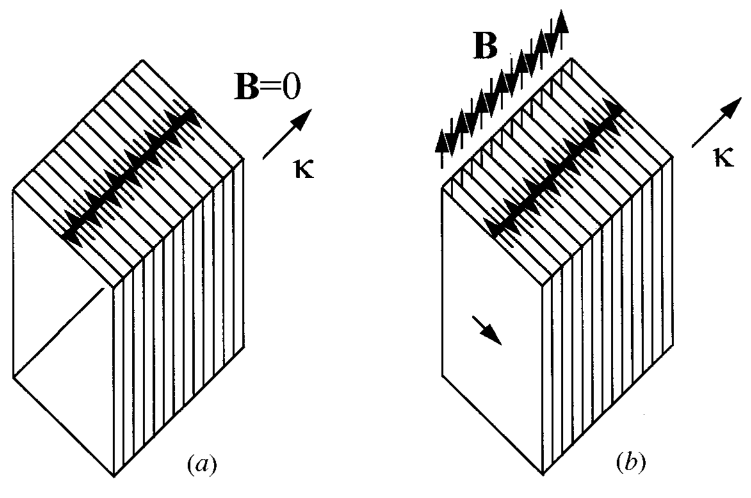

Fig. 1. Staggered magnetic dipole moments: (a) the result in zero magnetic induction if the directions of moment and staggering are the same; (b) the result in a staggered magnetic induction if the moment direction is perpendicular to the direction of staggering.

The matrix element can then be written

$$\left(\frac{2\pi\hbar^2}{m}\right)\langle s'q'|b(\boldsymbol{\kappa}) + \hat{\boldsymbol{A}}(\boldsymbol{\kappa})\,\boldsymbol{.}\,\hat{\boldsymbol{\sigma}}|qs\rangle\,, \tag{10}$$

in which $b(\boldsymbol{\kappa})$ is the Fourier transform of the scattering length density, and

$$\hat{\boldsymbol{A}}(\boldsymbol{\kappa}) = \sum_{\boldsymbol{R}} B(\boldsymbol{R})\,\hat{\boldsymbol{I}}(\boldsymbol{R})\,\mathrm{e}^{\mathrm{i}\boldsymbol{\kappa}\,\boldsymbol{.}\,\boldsymbol{R}} + \left(\frac{e\gamma}{\hbar c}\right)\hat{\boldsymbol{M}}_{\perp}(\boldsymbol{\kappa})\,, \tag{11}$$

with $\hat{\boldsymbol{M}}(\boldsymbol{\kappa})$ the Fourier transform of the total magnetic moment operator for the specimen.

3. Neutron Spin Transitions

At this point it is useful to consider the neutron spin transitions which are induced by the magnetic interaction. Among other uses a measurement of whether the neutron spin is flipped in a scattering event is an indication of whether the scattering event is magnetic or not.

If we evaluate the matrix element $U^{ss'}$ of equation (10) we have, for instance,

$$\begin{aligned} U^{+-} = (01)\left(\frac{2\pi\hbar^2}{m}\right)\langle q'|\left\{b + A_x(\boldsymbol{\kappa})\begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix} + A_y(\boldsymbol{\kappa})\begin{pmatrix}0 & -\mathrm{i}\\ \mathrm{i} & 0\end{pmatrix}\right. \\ \left. + A_z(\boldsymbol{\kappa})\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}\right\}|q\rangle\begin{pmatrix}1\\ 0\end{pmatrix}\,, \end{aligned} \tag{12}$$

with the matrices the x, y and z components of the Pauli spin matrix and the down and up spin states of the neutron indicated by the row and column matrices fore and aft. This gives

$$U^{+-} = \left(\frac{2\pi\hbar^2}{m}\right)\langle q'|[A_x(\boldsymbol{\kappa}) + \mathrm{i}\,A_y(\boldsymbol{\kappa})]|q\rangle\,. \tag{13}$$

Similarly

$$U^{++} = \left(\frac{2\pi\hbar^2}{m}\right)\langle q'|[b + A_z(\boldsymbol{\kappa})]|q\rangle\,. \tag{14}$$

It can be seen that only the x and y components of the nuclear spin and magnetic moment incorporated in $\boldsymbol{A}(\boldsymbol{\kappa})$ contribute to the neutron spin–flip amplitude, whereas the z components of the nuclear spin and the magnetic moment and the nuclear scattering amplitude contribute to the amplitude in which the neutron spin direction does not change. Put simply, components of nuclear spin and magnetic moment transverse to the neutron spin direction flip the neutron spin and components along the neutron spin direction and the nuclear scattering length preserve the neutron spin state. If the neutron spin direction is placed along the scattering vector, the cross section in which the neutron spin is not flipped consists only of the nuclear parts coming from the scattering length and the z component of the nuclear spin. The component of magnetic moment along the scattering vector does not scatter (9). All the magnetic scattering is then in the neutron spin–flip cross section and this is a useful device for isolating magnetic scattering.

A fuller analysis of neutron spin transitions involves transitions to neutron mixed spin states in which the final state spin precesses about the quantisation direction. This can be used to detect magnetic structures with a chiral component. Two alternative approaches to this problem can be found in Lovesey (1984) and Hicks (1995).

4. Elastic Scattering

The elastic scattering is obtained by requiring that the initial and final states of the scatterer, q and q', are the same and that the neutron's energy is unchanged. Putting $q' = q$ in the expressions in the previous section restricts the matrix element to be an expectation value. In particular the magnetic component is the expectation value of the magnetic moment $\langle \boldsymbol{M}_\perp(\boldsymbol{\kappa})\rangle$. Inserting this into (5) and integrating over the neutron energy change delta function gives

$$\frac{\mathrm{d}\sigma}{2\mathrm{d}\Omega} = \overline{\langle \hat{b}^*(\boldsymbol{\kappa})\rangle\, \langle \hat{b}(\boldsymbol{\kappa})\rangle} + \left(\frac{e\gamma}{\hbar c}\right)^2 \overline{\langle \hat{\boldsymbol{M}}^*_\perp(\boldsymbol{\kappa})\rangle\, \langle \hat{\boldsymbol{M}}_\perp(\boldsymbol{\kappa})\rangle} + \sum_{\boldsymbol{R},\boldsymbol{R}'} B(\boldsymbol{R})\, B(\boldsymbol{R}')\overline{\langle \hat{I}(\boldsymbol{R})\rangle\, \langle \hat{I}(\boldsymbol{R}')\rangle}\, \mathrm{e}^{\mathrm{i}(\boldsymbol{R}-\boldsymbol{R}')}\,, \tag{15}$$

in which the bar is the thermal average obtained from the sum over target state probabilities in (5). No cross terms between the three sources of scattering occur because they sum to zero in averaging over the neutron spin states in the unpolarised incident beams. The magnetic part of this is the thermal average of the square of the expectation value of the $\boldsymbol{\kappa}$th Fourier component of the magnetic moment. In particular, for a magnet with only long range order, magnetic scattering occurs only for particular values of $\boldsymbol{\kappa}$ equal to the reciprocal lattice vectors for the particular order. The intensity of the resulting Bragg peaks is then directly proportional to the thermally averaged square of the moment amplitude of the order.

5. Inelastic Scattering

One of the coincidences which makes neutron scattering a powerful tool in the study of condensed matter is that the energy of neutrons with wavelengths comparable to atomic spacings is similar to the energy of the thermal excitations of the matter being probed. Thus, the energy gained or lost by the neutrons in the interaction is generally a large fraction of their total energy and easily measurable. This results, in the magnetic case, in the ability to measure the magnetic excitations as a function of both wavevector and energy. The quantity measured is the absorptive or 'imaginary' part of the wavevector and energy dependent susceptibility; a most fundamental quantity in magnetism. We now explore the connection between the neutron cross section and the absorptive part of the generalised susceptibility.

The uniform frequency dependent susceptibility $\chi_{\alpha\alpha}(\omega)$ is defined as

$$M_\alpha(t) = \tfrac{1}{2} V[\chi_{\alpha\alpha}(\omega)\, H_{0\alpha}\, \mathrm{e}^{-\mathrm{i}\omega t} + \chi^*_{\alpha\alpha}(\omega)\, H^*_{0\alpha}\, \mathrm{e}^{\mathrm{i}\omega t}]\,, \tag{16}$$

where $M_\alpha(t)$ is the total moment resulting from the impressed oscillating field

$$H_\alpha(t) = \tfrac{1}{2}(H_{0\alpha}\, \mathrm{e}^{-\mathrm{i}\omega t} + H^*_{0\alpha}\, \mathrm{e}^{\mathrm{i}\omega t})\,, \tag{17}$$

and V is the specimen volume. The lag in response is described by the relative magnitudes of the real and imaginary parts of

$$\chi_{\alpha\alpha}(\omega) = \chi'_{\alpha\alpha}(\omega) + \mathrm{i}\, \chi''_{\alpha\alpha}(\omega)\,, \tag{18}$$

and the rate of energy gain in the specimen is

$$\frac{\mathrm{d}E}{\mathrm{d}t} = -M_\alpha \frac{\mathrm{d}H_\alpha}{\mathrm{d}t}\,, \tag{19}$$

so that the average rate of energy gain is

$$\overline{\frac{\mathrm{d}E}{\mathrm{d}t}} = \tfrac{1}{2} V H^*_{0\alpha}\, H_{0\alpha}\, \omega \chi''_{\alpha\alpha}(\omega)\,. \tag{20}$$

The power can also be calculated within first order perturbation theory as follows:

$$\overline{\frac{\mathrm{d}E}{\mathrm{d}t}} = \sum_q \sum_{q'} p_q\, W_{qq'}(E_{q'} - E_q)\,, \tag{21}$$

with p_q the probability that the system is in state q, and with $E_{q'}$ and E_q the energies of the two states. The transition probability is given by

$$W_{qq'} = \frac{\pi}{\hbar} H^*_{0\alpha}\, H_{0\alpha} |\langle q'|M_\alpha|q\rangle|^2 \delta(\hbar\omega - E_{q'} + E_q)\,. \tag{22}$$

But

$$\big|\langle q|M_\alpha|q'\rangle\big|^2 = \big|\langle q'|M_\alpha|q\rangle\big|^2\,, \tag{23}$$

and the nett rate in energy gain, i.e. absorption minus stimulated emission, is thus

$$\begin{aligned}\overline{\frac{\mathrm{d}E}{\mathrm{d}t}} &= \tfrac{1}{2} \sum_q \sum_{q\neq q'} \frac{\pi}{\hbar} H^*_{0\alpha}\, H_{0\alpha} \big|\langle q'|M_\alpha|q\rangle\big|^2 p_q \\ &\quad \times \left\{1 - \frac{\exp(-\beta E')}{\exp(-\beta E)}\right\}(E_{q'} - E_q)\, \delta(\hbar\omega - E_{q'} + E_q) \\ &= \tfrac{1}{2} \sum_q \sum_{q'\neq q} \pi H^*_{0\alpha}\, H_{0\alpha} \big|\langle q'|M_\alpha|q\rangle\big|^2 p_q\, \omega (1 - \mathrm{e}^{-\beta\hbar\omega})\, \delta(\hbar\omega - E_{q'} + E_q)\,, \end{aligned} \tag{24}$$

with $\frac{1}{2}$ compensating for double counting in the sum over the states, and omitting $q' = q$ because it involves no change in energy. Finally, by equating energy rates calculated in the two ways we have

$$\frac{\chi''_{\alpha\alpha}(\omega)}{\pi(1-\mathrm{e}^{-\beta\hbar\omega})} = \frac{1}{V}\sum_{q}\sum_{q'\neq q} p_q \left|\langle q'|M_\alpha|q\rangle\right|^2 \delta(\hbar\omega - E_{q'} + E_q)\,. \tag{25}$$

The cross sections may be obtained by recognising that $\langle q'|M_\alpha|q\rangle$ in this relation is part of the matrix element defined in (10) and can be related to the cross section through (5). The result for an unpolarised beam is

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E}\right)_{k,k'} = \frac{V}{\pi}\,\frac{k'}{k}\left(\frac{e\gamma}{\hbar c}\right)^2 \sum_\alpha \frac{\chi''_{\alpha\alpha}(\boldsymbol{\kappa},\,\omega)}{1-\mathrm{e}^{-\beta\hbar\omega}}\,, \tag{26}$$

with the sum over the two cartesian directions perpendicular to the scattering vector. In this way the inelastic neutron scattering cross section is directly proportional to the absorptive or 'imaginary' part of the susceptibility.

This relationship can be made really useful for a comparison of the neutron cross section with a bulk susceptibility measurement by use of the Kramers–Kronig relations which connect the real and imaginary parts of the susceptibility. The generalised susceptibility may be written

$$\chi(\boldsymbol{\kappa},\,\omega) = \int_0^\infty \mathrm{d}t\; X(\boldsymbol{\kappa},\,t)\mathrm{e}^{\mathrm{i}\omega t}\,, \tag{27}$$

in which $X(\boldsymbol{\kappa},\,t)$ is defined only for $t > 0$. Then

$$\chi'(\boldsymbol{\kappa},\,\omega) = \int_0^\infty \mathrm{d}t\; X(\boldsymbol{\kappa},\,t)\cos\omega t\,, \qquad \chi''(\boldsymbol{\kappa},\,\omega) = \int_0^\infty \mathrm{d}t\; X(\boldsymbol{\kappa},\,t)\sin\omega t\,, \tag{28}$$

$$\int_{-\infty}^\infty \mathrm{d}\omega\;\frac{\chi''(\boldsymbol{\kappa},\,\omega)}{\omega} = \int_0^\infty \mathrm{d}t\; X(\boldsymbol{\kappa},\,t)\int_{-\infty}^\infty \mathrm{d}\omega\;\frac{\sin\omega t}{\omega} = \pi\chi'(\boldsymbol{\kappa},\,\omega=0)\,, \tag{29}$$

which is a special case of the Kramers–Kronig relationships. If we now integrate the cross section over energy we have

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \int_{-\infty}^\infty \hbar\,\mathrm{d}\omega\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right) \approx \int_{-\infty}^\infty \hbar\,\mathrm{d}\omega\left(\frac{e\gamma}{\hbar c}\right)^2 \frac{V}{\pi\beta\hbar\omega}\sum_\alpha \chi''_{\alpha\alpha}(\boldsymbol{\kappa},\,\omega)\,, \tag{30}$$

and for $\hbar\omega \ll 1/\beta$ and $k' \approx k$,

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = V\left(\frac{e\gamma}{\hbar c}\right)^2 k_{\mathrm{B}}\,T\sum_\alpha \chi'_{\alpha\alpha}(\boldsymbol{\kappa},\,0)\,,$$

using the Kramers–Kronig relationship derived above. This is the quasielastic approximation for the magnetic cross section and is useful for comparison of the neutron cross section with bulk susceptibility measurements. As can be seen, subject to the conditions above, the energy integrated cross section is directly related to the zero frequency or truly dc susceptibility which for $\boldsymbol{\kappa} = 0$ is measured on a magnetometer. This can be important to check whether a magnetometer is measuring the true dc susceptibility which will be further discussed in the following paper (Hicks 1997, present issue p. 1119).

6. Conclusions

The purpose of this paper was to demonstrate the close relationship between the magnetic neutron scattering cross section and the magnetisation for elastic scattering and the susceptibility for inelastic scattering. From a practical, experimental point of view the least this analysis does is to allow the experimentalist to include an extra $\boldsymbol{\kappa} = 0$ point in his/her neutron scattering data. The reward, however, is more than this. It permits the experimentalist an extra insight into his/her neutron data and suggests modelling which comes from the treatment of magnetisations and susceptibilities. This will be illustrated with concrete and recent neutron data in the following paper (Hicks 1997).

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