The scattering of polarized neutrons by a magnetic material

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Abstract We address the properties of the cross-section for the magnetic scattering of polarized neutrons. Results are given for cross-sections that describe total scattering, diffraction (Bragg scattering) and inelastic scattering events. The properties discussed flow from basic principles, namely the condition of detailed balance in scattering and the invariance of the sample's properties to a simultaneous reversal in the directions of time and the polarity of the magnetization. Attention is given to features that can arise with a non-collinear arrangement of the atomic magnetic moments and anisotropic interactions between the moments.

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1. **Introduction**

The scattering of radiation by a material is a very powerful technique by which to recover information on the properties of the material at an atomic level of detail. For magnetic properties the unrivalled radiation is a beam of neutrons. The information available from an analysis of the scattered radiation includes the orientation and location of the atomic magnetic moments, and the frequency spectra of excitations and fluctuations.

The neutron cross-sections for elastic and inelastic events have been derived and investigated by many authors over a period of several decades [1]. With the growing interest in properties of complex magnetic materials, some possessing anisotropic interactions and non-collinear moment configurations, it is, perhaps, timely to address the cross-sections for elastic, inelastic and total scattering of polarized neutrons. We gather properties of these cross-sections that flow from the condition of detailed balance in scattering events, and time-reversal invariance applied to the properties of the sample. We choose to articulate in the language of correlation functions and the van Hove spin response function. Also, we have occasion to call on the theory of linear response.

2. **Identities**

The thermal average value of Q, a quantum-mechanical operator, is denoted by angular brackets. For an arbitrary operator Q,

$$\langle Q \rangle^* = (\mathrm{Tr.}\rho Q)^* = \langle Q^+ \rangle, \tag{2.1}$$

where Q^+ is the Hermitian conjugate of Q, and ρ is the density matrix that describes the equilibrium properties of the sample. The identity (2.1) guarantees that the cross-section is purely real, as it must be.

Due to the principle of micro-reversibility, the sample's properties are not changed by the simultaneous reversal of the direction of time and the polarity of the magnetization. Let us denote by Q' the operator formed from Q by the operation of time reversal alone. By way of an example, let us consider $Q = \exp(\mathrm{i}\mathbf{k} \cdot \mathbf{R})\mathbf{p}$, where $\mathbf{p}$ is the momentum operator conjugate to the position operator $\mathbf{R}$. One has, $Q' = -\exp(-\mathrm{i}\mathbf{k} \cdot \mathbf{R})\mathbf{p}$. This result follows because $\mathbf{p}$ and $\mathbf{R}$, respectively, are odd and even with respect to the operation of time reversal, and this operation also changes i to −i, i.e. it includes complex conjugation.

If we denote the magnetic field acting on the sample by $\mathbf{H}$, time-reversal invariance at the atomic level requires [2, 3] the identity,

$$\langle Q \rangle_{\mathbf{H}} = \left\langle \{Q'\}^{+} \right\rangle_{-\mathbf{H}}. \tag{2.2}$$

Taking Q to be the total magnetic moment of the sample, $\mathbf{M}$, (2.2) gives, $\langle \mathbf{M} \rangle_{\mathbf{H}} = -\langle \mathbf{M} \rangle_{-\mathbf{H}}$, which provides the relation between $\langle \mathbf{M} \rangle$ and $\mathbf{H}$. If the sample is paramagnetic the field-induced magnetization vanishes as the field is reduced to zero. For a state of spontaneous magnetization $\langle \mathbf{M} \rangle$ is non-zero in the limit $\mathbf{H} = 0$, and the polarity of the spontaneous magnetization is determined by the polarity of $\mathbf{H}$.

For the third, and last, identity that is of interest in connection with our discussion of the cross-section we consider $Q = A(0)B(t)$, where $A(t = 0)$ and $B(t)$ are Heisenberg operators derived from arbitrary A and B. Quantum properties make $\langle A(0)B(t) \rangle$ and $\langle B(0)A(t) \rangle$ different. One finds,

$$\langle A(0)B(t) \rangle = \langle B(0)A(\mathrm{i}\hbar\beta - t) \rangle, \tag{2.3}$$

where $\beta = 1/k_{\mathrm{B}} T$ and T is the temperature. Often, (2.3) is referred to as the condition of detailed balance; in fact, one of many guises that the condition can adopt.

3. Cross-section

The interaction with the nuclei is described by the operator, $b\, n(\mathbf{k})$. Here, b is the average value of the scattering length, $\mathbf{k}$ is the scattering wave vector and $n(\mathbf{k}) = \sum \exp(\mathrm{i}\mathbf{k} \cdot \mathbf{R}_j)$, where the sum is over all nuclei. For the interaction with atomic moments we use the operator [4],

$$\mathbf{T}(\mathbf{k}) = \tfrac{1}{2} g F(\mathbf{k}) \sum_a \exp(\mathrm{i}\mathbf{k}\cdot\mathbf{R}_a)\{\hat{\mathbf{k}} \text{ x } (\mathbf{S}_a \text{ x } \hat{\mathbf{k}})\}.$$

In this expression, $\hat{\mathbf{k}} = \mathbf{k}/k$, and g, $F(\mathbf{k})$, $\mathbf{R}$ and $\mathbf{S}$ are, respectively, the gyromagnetic factor, atomic form factor, position and spin operator of one of the identical magnetic atoms. We will assume that $F(\mathbf{k}) = F(-\mathbf{k})$. The foregoing expression for the magnetic interaction operator is approximate, to the extent that orbital angular momentum is not treated in full. Instead, orbital angular momentum is only partially included by allowing the gyromagnetic factor to depart from its pure-spin value.

If the primary beam of neutrons carries a polarization $\mathbf{P}$ the magnetic cross-section is proportional to,

$$\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} \mathrm{d}t \, \exp(-\mathrm{i}\omega t)\left\{r_0^2\left\langle \mathbf{T}^+ \cdot \mathbf{T}(t)\right\rangle + r_0 b\left\langle n^+ \mathbf{P}\cdot\mathbf{T}(t)\right\rangle + r_0 b\left\langle \mathbf{P}\cdot\mathbf{T}^+ n(t)\right\rangle + \mathrm{i} r_0^2 \mathbf{P}\cdot\left\langle \mathbf{T}^+ \text{ x } \mathbf{T}(t)\right\rangle\right\}. \quad (3.1)$$

Here, $\hbar\omega$ is the energy transferred to the sample and $r_0 = -0.54.\ 10^{-12}$cm.

In later parts of the paper we are led to consider the signals observed at the geometrical settings that give scattering wave vectors k and - k,

4. Interference scattering

If we introduce the mixed response function,

$$\mathbf{Z}_{\mathbf{H}}(\mathbf{k},\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} \mathrm{d}t \, \exp(-\mathrm{i}\omega t)\left\langle n(-\mathbf{k},0)\mathbf{T}(\mathbf{k},t)\right\rangle_{\mathbf{H}}, \quad (4.1)$$

the interference contribution to (3.1), induced by the polarization, is proportional to,

$$\mathbf{P}\cdot\left\{\mathbf{Z}_{\mathbf{H}}(\mathbf{k},\omega) - \mathbf{Z}_{-\mathbf{H}}(-\mathbf{k},\omega)\right\} \quad (4.2)$$

$$= \mathbf{P} \cdot \left\{ \mathbf{Z_H}(\mathbf{k},\omega) + \mathbf{Z_H}(-\mathbf{k},-\omega)\exp(\hbar\omega\beta) \right\}. \qquad (4.3)$$

The results (4.2) and (4.3) follow from (2.2) and (2.3), respectively, which give,

$$\mathbf{Z_H}(\mathbf{k},\omega) = -\,\mathbf{Z}_{-\mathbf{H}}(\mathbf{k},-\omega)\exp(\hbar\omega\beta). \qquad (4.4)$$

Notice we do not assume that the scattered signal is the same at **k** and – **k**. A difference in the signals can arise if the atomic moments occupy sites that are not centres of inversion symmetry, or the magnetic long-range order is non-collinear.

The result (4.2) is particularly interesting. It shows that, the interference scattering is odd with respect to a simultaneous change in the sign of **k** and the polarity of the magnetization. Hence, for some magnetic materials the interference scattering does not merely change sign on changing the polarity of the magnetization.

The results (4.2) and (4.3) apply to both inelastic and elastic ($\omega = 0$) scattering events. For Bragg diffraction (4.3) tells us that the interference scattering is always the same at **k** and – **k**, for a given value of **H**.

5. Purely magnetic scattering

It is useful to introduce a van Hove response function for the spin operaters,

$$S^{\alpha\beta}(\mathbf{k},\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} \mathrm{d}t\, \exp(-\mathrm{i}\omega t)\sum_{a,b} \exp\{\mathrm{i}\mathbf{k}\cdot(\mathbf{R}_b - \mathbf{R}_a)\}\langle S_a^{\alpha}(0) S_b^{\beta}(t)\rangle = \left\{S^{\beta\alpha}(\mathbf{k},\omega)\right\}^*, \qquad (5.1)$$

and the second equality follows from (2.1). In terms of the van Hove function the purely magnetic part of (3.1) is proportional to,

$$\sum_{a,b} S^{\alpha\beta}(\mathbf{k},\omega)\left[(\delta_{\alpha\beta} - \hat{\mathbf{k}}_\alpha \hat{\mathbf{k}}_\beta) + \mathrm{i}G^{\alpha\beta}(\mathbf{P},\mathbf{k})\right] = \sum_{\alpha,\beta}\left[(\delta_{\alpha\beta} - \hat{\mathbf{k}}_\alpha \hat{\mathbf{k}}_\beta) A^{\alpha\beta}(\mathbf{k},\omega) + G^{\alpha\beta}(\mathbf{P},\mathbf{k}) B^{\alpha\beta}(\mathbf{k},\omega)\right].$$

(5.2)

Here, $G^{\alpha\beta}(\mathbf{P},\mathbf{k}) = -G^{\beta\alpha}(\mathbf{P},\mathbf{k}) = (\hat{\mathbf{k}}\cdot\mathbf{P})\sum \varepsilon^{\alpha\beta\gamma}\hat{k}_{\gamma}$ where $\varepsilon^{\alpha\beta\gamma}$ is the completely antisymmetric tensor with three Cartesian indices, and we have introduced the symmetric and antisymmetric combinations,

$$A^{\alpha\beta}(\mathbf{k},\omega) = \tfrac{1}{2}\{S^{\alpha\beta}(\mathbf{k},\omega) + S^{\beta\alpha}(\mathbf{k},\omega)\} = \mathrm{Re.}\, S^{\alpha\beta}(\mathbf{k},\omega)\,, \tag{5.3}$$

and

$$B^{\alpha\beta}(\mathbf{k},\omega) = \tfrac{\mathrm{i}}{2}\{S^{\alpha\beta}(\mathbf{k},\omega) - S^{\beta\alpha}(\mathbf{k},\omega)\} = -\,\mathrm{Im.}\, S^{\alpha\beta}(\mathbf{k},\omega). \tag{5.4}$$

The results (5.3) and (5.4) show that the polarization-dependent and polarization-independent parts of the purely magnetic cross-section measure quite different magnetic correlations. We pursue this feature of the cross-section by expressing $A^{\alpha\beta}$ and $B^{\alpha\beta}$ in terms of the generalized spin-susceptibility.

It is most convenient to start from the spin response-function, defined as the spatial Fourier transform of a commutation of two spin operators,

$$\phi^{\alpha\beta}(\mathbf{k},t) = \tfrac{\mathrm{i}}{\hbar}\sum_{a,b} \exp\{\mathrm{i}\mathbf{k}\cdot(\mathbf{R}_a - \mathbf{R}_b)\}\left\langle\left[S_a^{\alpha}(t), S_b^{\beta}(0)\right]\right\rangle, \tag{5.5}$$

and it satisfies the identity, $\phi^{\beta\alpha}(\mathbf{k},t) = -\{\phi^{\alpha\beta}(\mathbf{k},-t)\}^{*}$. The generalized susceptibility is the Laplace transform of the spin response-function, viz.,

$$\chi^{\alpha\beta}[\mathbf{k},\omega] = -\int_0^{\infty} \mathrm{d}t\; \phi^{\alpha\beta}(\mathbf{k},t)\exp\{-t(\eta + \mathrm{i}\omega)\}$$

where $\eta \to 0$ is understood. For the van Hove response function (5.1) one finds,

$$S^{\alpha\beta}(\mathbf{k},\omega) = \frac{i}{2\pi}\{1+n(\omega)\}\int_{-\infty}^{\infty} dt\ \phi^{\beta\alpha}(\mathbf{k},t)\exp(-i\omega t), \qquad (5.7)$$

with $n(\omega) = \{\exp(\hbar\omega\beta) - 1\}^{-1}$.

The following relations are also quite easy to obtain,

$$A^{\alpha\beta}(\mathbf{k},\omega) = \frac{1}{2\pi}\{1+n(\omega)\}\mathrm{Im.}\left\{\chi^{\alpha\beta}[\mathbf{k},\omega] + \chi^{\beta\alpha}[\mathbf{k},\omega]\right\} \qquad (5.8)$$

and,

$$B^{\alpha\beta}(\mathbf{k},\omega) = \frac{-1}{2\pi}\{1+n(\omega)\}\mathrm{Re.}\left\{\chi^{\alpha\beta}[\mathbf{k},\omega] - \chi^{\beta\alpha}[\mathbf{k},\omega]\right\}. \qquad (5.9)$$

From this pair of relations it follows that the first and last contributions to the cross-section (3.1), respectively, are proportional to Im. $\chi^{\alpha\beta}$ $[\mathbf{k},\omega]$ and Re. $\chi^{\alpha\beta}$ $[\mathbf{k},\omega]$. In consequence, an experiment made with a beam of polarized neutrons in principle enables the analytic function $\chi^{\alpha\beta}$ $[\mathbf{k},\omega]$ to be measured. Let us mention that, while the real and imaginary parts of $\chi^{\alpha\beta}$ $[\mathbf{k},\omega]$ do not satisfy a simple dispersion relation, the linear combinations of the susceptibility in $A^{\alpha\beta}$ and $B^{\alpha\beta}$ do satisfy such a relation.

The total scattering generated by the antisymmetric combination (5.4) is,

$$\int_{-\infty}^{\infty} d\omega\ B^{\alpha\beta}(\mathbf{k},\omega) = -\tfrac{1}{2}\sum_{a}\sum_{\gamma}\varepsilon^{\alpha\beta\gamma}\left\langle S_a^{\gamma}\right\rangle + \sum_{a,b}\sin\left\{\mathbf{k}\cdot(\mathbf{R}_a - \mathbf{R}_b)\right\}\left\langle S_a^{\alpha}S_b^{\beta}\right\rangle. \qquad (5.10)$$

The first term in (5.10) is a direct consequence of the commutation relation for $S_a^{\alpha}S_b^{\beta}$, and it is evidently zero for a magnetic material with no long-range order. One finds the first term contributes to the cross-section for total scattering a factor, $-(\hat{\mathbf{k}}\cdot\mathbf{P})(\hat{\mathbf{k}}\cdot\mathbf{M})$. Hence, the total scattering contains the projection of the magnetization on $\mathbf{k}$. This finding contrasts with the cross-section for Bragg diffraction (a time averaged process whereas the total scattering is an instantaneous process), which contains components of the spatial Fourier transform of the magnetization that are perpendicular to **k**.

The second term in (5.10) is zero for a material in which the magnetic ions occupy sites that are centro-symmetric for, in this instance, the two-spin correlation function depends only on $|\mathbf{R}_a - \mathbf{R}_b|$. In addition, when there is no long-range order and $\alpha \neq \beta$ the correlation function $\langle S_a^\alpha S_b^\beta \rangle$ is zero unless the spin Hamiltonian contains an anisotropic interaction, e.g. the Dzyaloshinsky-Moriya exchange interaction. We conclude that, in the absence of long-range magnetic order in the target sample the total scattering generated by $B^{\alpha\beta}$ most likely is zero. Exceptions are various novel materials, like a dimerized chain with anisotropic exchange interactions. The second term in (5.10) contributes to the cross-section for total scattering a factor,

$$(\hat{\mathbf{k}} \cdot \mathbf{P}) \sum_{a,b} \sin\{\mathbf{k} \cdot (\mathbf{R}_a - \mathbf{R}_b\} \, \hat{\mathbf{k}} \cdot \langle \mathbf{S}_a \text{ x } \mathbf{S}_b \rangle. \tag{5.11}$$

It is interesting to observe that this contains the projection of **k** on a chiral order-parameter.

The following relations are obtained from the identities in section 3;

$$B_{\mathbf{H}}^{\alpha\beta}(\mathbf{k},\omega) = \tfrac{\mathrm{i}}{2}\left\{S_{\mathbf{H}}^{\alpha\beta}(\mathbf{k},\omega) - S_{-\mathbf{H}}^{\alpha\beta}(-\mathbf{k},\omega)\right\}, \tag{5.12}$$

from which,

$$B_{\mathbf{H}}^{\alpha\beta}(\mathbf{k},\omega) + B_{-\mathbf{H}}^{\alpha\beta}(-\mathbf{k},\omega) = 0. \tag{5.13}$$

And,

$$B^{\alpha\beta}(\mathbf{k},\omega) = -B^{\alpha\beta}(-\mathbf{k},-\omega)\exp(\hbar\omega\beta), \tag{5.14}$$

which evaluated for $\omega = 0$ gives,

$$B^{\alpha\beta}(\mathbf{k},0) + B^{\alpha\beta}(-\mathbf{k},0) = 0. \tag{5.15}$$

Several aspects of these relations merit comment.

In light of (5.12), we can view $B_{\mathbf{H}}^{\alpha\beta}$ as a measure of the chiral signature of excitations. Consider, for example, **H** aligned with the *z*-axis. In this instance, $S_{\mathbf{H}}^{xy}(\mathbf{k},\omega)$ describes excitations with circular polarization, the corresponding helicity is parallel to the *z*-axis, and $B_{\mathbf{H}}^{xy}(\mathbf{k},\omega)$ is a difference between the left and right-handed helicity states. The relation (5.13) shows that, $B_{\mathbf{H}}^{\alpha\beta}$ is possibly different from zero even in the absence of a preferred magnetic axis in the target sample, given the ions occupy non-centro-symmetric sites and $\mathbf{k} \neq 0$. A necessary condition for this to occur is that the Hamiltonian of the spins is anisotropic with respect to the components of the spins, e.g. $S_{\mathbf{H}}^{xy}(\mathbf{k},\omega) \neq 0$ for $\mathbf{H} = 0$ and no spontaneous ordering of the moments. Lastly, from (5.15) we conclude that, if the response function is independent of the sign of **k**, so the response is spatially isotropic, the antisymmetric part of it is zero for $\omega = 0$.

6. Conclusions

In our treatment of the subject we have concentrated primarily on those aspects of the scattering of neutrons which are governed by the polarization **P** of the neutron. The essential parts for the contributions to the cross-section, couched in the language of linear response theory, are represented by a van Hove response function for spin operators or a mixed response function for purely magnetic or interference scattering, respectively.

The main thrust of our discussion is directed at the consequences of two symmetry principles impressed on the correlation and response functions, namely the condition of detailed balance and the time reversal invariance at the microscopic level. We take care not to limit our treatment to such situations (samples) where the scattering remains the same when the direction of the scattering vector is reversed. The theory remains, thus, applicable to cases where atomic moments are not at sites with inversion symmetry or where the sample has non-collinear long-range order.

In general the response functions for a certain sample (and experiment) will depend on the direction of the wave vector transfer **k**, the applied field **H**, and the energy transferred in the scattering process. We note that, the properties and (inter)relations flowing from the aforementioned symmetry conditions force, in some cases, a reversal of the applied field together with the reversal of **k**, in order to arrive at the appropriate combinations for the scattering function. The interference scattering is, consequently, described by the projection of $\left\{\mathbf{Z}_{\mathbf{H}}(\mathbf{k},\omega)-\mathbf{Z}_{-\mathbf{H}}(-\mathbf{k},\omega)\right\}$ onto the polarization **P,** an expression antisymmetric with respect to a simultaneous reversal of both **k** and **H.** In the case of elastic scattering, with ω = 0, the relevant expression can be rewritten as $\left\{\mathbf{Z}_{\mathbf{H}}(\mathbf{k},0)+\mathbf{Z}_{\mathbf{H}}(-\mathbf{k},0)\right\}$, thus showing the symmetry of Bragg scattering with respect to the reversal from **k** to **–k**.

Considering purely magnetic scattering, we find the polarization-dependent contribution to be proportional to $\mathrm{Re.}\left\{\chi^{\alpha\beta}\left[\mathbf{k},\omega\right]-\chi^{\beta\alpha}\left[\mathbf{k},\omega\right]\right\}$ or $\frac{i}{2}\left\{S^{\alpha\beta}_{\mathbf{H}}(\mathbf{k},\omega)-S^{\alpha\beta}_{-\mathbf{H}}(-\mathbf{k},\omega)\right\}$, if the generalized susceptibility or the van Hove response function is used, respectively.
And, therefore, …

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References

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