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Magnetically scattered polarized neutrons: static and dynamic features of the sample

Stephen W Lovesey and Greg I Watson
ISIS Facility, Rutherford Appleton Laboratory, Oxfordshire OX11 0QX, UK

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Abstract. An investigation of the cross-section for polarized neutrons scattered by a magnetic material is reported. Included in the investigation are the cross-sections for elastic (Bragg), total, and inelastic scattering processes. Special attention is paid to the information available on the chiral character of magnetic states. It is possible that the magnetic response function (also known as the spin van Hove function) is spatially anisotropic, in which case the observed signals are different for the scattering wave vectors $\boldsymbol{k}$ and $-\boldsymbol{k}$. This situation can arise with materials in which the magnetic ions are not at centres of inversion symmetry, and when the magnetic order is not collinear, e.g. a helimagnet.

1. Introduction

The perspicacity to magnetic properties of materials that is special in using the technique of neutron scattering is realized to the full with methods which exploit the polarization of neutron beams [1]. In Bragg-diffraction experiments it is today almost routine practice to employ polarized-beam methods, most commonly polarization of the primary beam to induce an interference between the magnetic and nuclear scattering amplitudes. The interference term in the cross-section, being linear in the magnetic amplitude, offers enhanced sensitivity and the sign of the amplitude. The empirical information on the magnetic properties features both the configuration of the moments and the spatial distribution of the magnetization. For example, in a recent experiment, using a reflectometer, the first of these two features has been nicely exploited to find the chirality of layers of magnetic material [2].

Because of the less than perfect efficiency of polarization devices, currently polarization methods are not common practice in measurements of inelastically scattered neutrons where the signals involved are quite small compared to Bragg intensities. Even so, a large number of successful experiments have been reported in the past three decades [3–5]. In most cases a polarized beam is used to effect a separation of the nuclear (lattice vibrations) and magnetic contributions to scattered signals.

The large diversity of materials of current interest, like spin ladders, dimerized chains, and molecular magnets, makes it timely to take a fresh look at the information that can be gathered about a sample of a magnetic material by scattering a beam of polarized neutrons from it. Here, we discuss the appropriate cross-section for the scattering process in terms of a van Hove response function for spin operators. (It is assumed that the magnetic ions are identical.) For theoretical investigations, it might be useful to derive the response function from an auxiliary function better suited than the response function itself for calculation, e.g. a Green function or Kubo's relaxation function.

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We present a number of new results that should be useful in the interpretation and design of experiments looking at Bragg diffraction, total scattering or inelastic scattering. In the discussion, attention is given to effects that can occur in materials that do not support a long-range magnetic order, and possess anisotropic exchange interactions and non-centrosymmetric sites for the magnetic ions. When discussing ordered states we allow for non-collinear configurations of the moments, found in a weak ferromagnet and a helimagnet. Another feature of our work, hopefully, is a perspicuous account of the chirality of excitations as seen in the cross-section for inelastically scattered polarized neutrons.

We make good shortcomings in a recent paper [6]. The authors of [6] set aside significant previous experimental work, and their theoretical treatment is incomplete to the extent that, when corrected, its validity is restricted to scattering which is spatially isotropic, so it excludes, for example, non-collinear configurations.

Most of our findings are reported in section 3, following a review of the cross-section for polarized neutrons scattered by a magnetic material. Conclusions are gathered in section 4.

2. The cross-section for scattering a polarized beam

Perhaps the most compact expression for the cross-section for neutrons scattered by unpaired electrons comes from expressing the magnetic interaction as

$$V(\boldsymbol{k}) = \boldsymbol{\sigma} \cdot \boldsymbol{T}(\boldsymbol{k}) \tag{2.1}$$

where $\boldsymbol{\sigma}$ is twice the value of the spin of the neutron and $\boldsymbol{k} = \boldsymbol{q} - \boldsymbol{q}'$ is the change in the wave vector of the neutron caused by scattering. A useful approximation to the atomic variable $\boldsymbol{T}(\boldsymbol{k})$, in which the orbital contribution to the magnetic moment is described by a gyromagnetic factor different from the pure-spin value, is

$$\boldsymbol{T}(\boldsymbol{k}) = \frac{1}{2} \sum_a g_a F_a(\boldsymbol{k}) \exp(\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{R}_a)\{\hat{\boldsymbol{k}} \times (\boldsymbol{S}_a \times \hat{\boldsymbol{k}})\}. \tag{2.2}$$

Here, the unit vector $\hat{\boldsymbol{k}} = \boldsymbol{k}/k$, and g_a, $F_a(\boldsymbol{k})$, $\boldsymbol{R}_a$, and $\boldsymbol{S}_a$ are, respectively, the gyromagnetic factor, atomic form factor, position and spin operator of the ion labelled by the index a. If the primary beam of neutrons has a polarization $\boldsymbol{P}$, the cross-section is proportional to [7, 8]

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}t\ \exp(-\mathrm{i}\omega t)\{\langle \boldsymbol{T}^+(\boldsymbol{k}) \cdot \boldsymbol{T}(\boldsymbol{k}, t)\rangle + \mathrm{i}\boldsymbol{P} \cdot \langle \boldsymbol{T}^+(\boldsymbol{k}) \times \boldsymbol{T}(\boldsymbol{k}, t)\rangle\}. \tag{2.3}$$

In this expression, $\overline{h} = 1$, $\omega = (q^2 - q'^2)/2m$ is the energy transferred from the beam to the sample, and $\langle \cdots \rangle$ denotes the thermal average of the enclosed Heisenberg operator.

The polarization of the secondary beam of neutrons contains a term proportional to $\langle \boldsymbol{T}^+ \times \boldsymbol{T}\rangle$. This term is responsible for creating polarization from an unpolarized ($\boldsymbol{P} = 0$) primary beam.

The cross-section that describes Bragg diffraction, which is a strictly elastic scattering process, is found from (2.3) by taking the limit $t \to \infty$ in the correlation functions. The result

$$\langle \boldsymbol{T}^+(\boldsymbol{k}) \cdot \boldsymbol{T}(\boldsymbol{k}, t = \infty)\rangle = |\langle \boldsymbol{T}(\boldsymbol{k})\rangle|^2 \tag{2.4}$$

is correct for a bulk sample, and the polarization-dependent contribution in (2.3) is treated in a similar manner [8]. Not included in the foregoing discussion is the interference between nuclear and magnetic scattering amplitudes induced by polarization in the primary beam.

For materials with overlapping nuclear and magnetic Bragg reflections, like ferromagnets, the interference affords a valuable method for obtaining accurate data.

Henceforth, we consider magnetic ions that have common values for the gyromagnetic and atomic form factors. In this case, for example,

$$\langle \boldsymbol{T}^{+}(\boldsymbol{k}) \cdot \boldsymbol{T}(\boldsymbol{k}, t)\rangle = \left\{\frac{1}{2} g F(\boldsymbol{k})\right\}^2 \sum_{\alpha,\beta} (\delta_{\alpha\beta} - \hat{k}_\alpha \hat{k}_\beta) \sum_{a,b} \exp\{\mathrm{i}\boldsymbol{k} \cdot (\boldsymbol{R}_b - \boldsymbol{R}_a)\} \langle S_a^\alpha S_b^\beta(t)\rangle \tag{2.5}$$

where α and β label Cartesian components. It will prove convenient to use a van Hove response function defined by

$$S^{\alpha\beta}(\boldsymbol{k}, \omega) = \frac{1}{2\pi N} \int_{-\infty}^{\infty} \mathrm{d}t \ \exp(-\mathrm{i}\omega t) \sum_{a,b} \exp\{\mathrm{i}\boldsymbol{k} \cdot (\boldsymbol{R}_b - \boldsymbol{R}_a)\} \langle S_a^\alpha S_b^\beta(t)\rangle \tag{2.6}$$

where N is the number of identical magnetic ions. The cross-section (2.3) normalized by the number of ions is proportional to

$$\left\{\frac{1}{2} g F(\boldsymbol{k})\right\}^2 \sum_{\alpha,\beta} S^{\alpha\beta}(\boldsymbol{k}, \omega)[(\delta_{\alpha\beta} - \hat{k}_\alpha \hat{k}_\beta) + \mathrm{i}G^{\alpha\beta}(\boldsymbol{P}, \boldsymbol{k})] \tag{2.7}$$

and, on using equation (10.81) from [8],

$$G^{\alpha\beta}(\boldsymbol{P}, \boldsymbol{k}) = (\hat{\boldsymbol{k}} \cdot \boldsymbol{P}) \sum_{\gamma} \varepsilon^{\alpha\beta\gamma} \hat{k}_\gamma = G^{\alpha\beta}(\boldsymbol{P}, -\boldsymbol{k}) \tag{2.8}$$

where the antisymmetric unit tensor of rank three (also called the unit axial tensor) gives for the vector product of $\boldsymbol{k}$ and $\boldsymbol{P}$, say,

$$(\hat{\boldsymbol{k}} \times \boldsymbol{P})_\alpha = \sum_{\beta,\gamma} \varepsilon^{\alpha\beta\gamma} \hat{k}_\beta P_\gamma .$$

In (2.7) we note that the first term inside the square brackets, which is independent of the polarization, is even with respect to an interchange of the Cartesian labels α and β, while $G^{\alpha\beta}$ is odd with respect to these labels ($G^{\alpha\beta}$ is actually the same as the corresponding quantity in equation (A3) of [6], where the expression is not reduced to the simple form shown in (2.8)). This observation suggests that it is sensible to write

$$S^{\alpha\beta}(\boldsymbol{k}, \omega) = A^{\alpha\beta}(\boldsymbol{k}, \omega) - \mathrm{i}B^{\alpha\beta}(\boldsymbol{k}, \omega) \tag{2.9}$$

where

$$A^{\alpha\beta}(\boldsymbol{k}, \omega) = \frac{1}{2}\{S^{\alpha\beta}(\boldsymbol{k}, \omega) + S^{\beta\alpha}(\boldsymbol{k}, \omega)\} = A^{\beta\alpha}(\boldsymbol{k}, \omega) \tag{2.10}$$

and

$$B^{\alpha\beta}(\boldsymbol{k}, \omega) = \frac{\mathrm{i}}{2}\{S^{\alpha\beta}(\boldsymbol{k}, \omega) - S^{\beta\alpha}(\boldsymbol{k}, \omega)\} = -B^{\beta\alpha}(\boldsymbol{k}, \omega). \tag{2.11}$$

Inserting these expressions in (2.7), the latter reduces to

$$\left\{\frac{1}{2} g F(\boldsymbol{k})\right\}^2 \sum_{\alpha,\beta} [(\delta_{\alpha\beta} - \hat{k}_\alpha \hat{k}_\beta) A^{\alpha\beta}(\boldsymbol{k}, \omega) + G^{\alpha\beta}(\boldsymbol{P}, \boldsymbol{k}) B^{\alpha\beta}(\boldsymbol{k}, \omega)]. \tag{2.12}$$

There is something to be said in favour of introducing a vector $\boldsymbol{B}$ defined in terms of $B^{\alpha\beta}$ by

$$B_\gamma = \frac{1}{2} \sum_{\alpha,\beta} B^{\alpha\beta} \varepsilon^{\alpha\beta\gamma} \tag{2.13}$$

from which we get

$$B_x = B^{yz} \qquad B_y = B^{zx} \qquad B_z = B^{xy}. \tag{2.14}$$

Using the standard identity

$$\sum_{\alpha,\beta} \varepsilon^{\alpha\beta\gamma}\varepsilon^{\alpha\beta\gamma'} = 2\delta_{\gamma\gamma'}$$

and the definitions (2.8) and (2.13), one finds

$$\sum_{\alpha,\beta} G^{\alpha\beta}B^{\alpha\beta} = 2(\hat{\boldsymbol{k}}\cdot\boldsymbol{P})(\hat{\boldsymbol{k}}\cdot\boldsymbol{B}). \tag{2.15}$$

Thus, the contribution to the cross-section induced by the polarization is proportional to the product of the projection of $\boldsymbol{P}$ and the projection of $\boldsymbol{B}$ on the scattering vector.

Since $B^{\alpha\beta}$ is constructed from the difference of the products of two spin operators with inverted order, one anticipates that its actual value hinges on the commutation relation, or Lie algebra, for spin operators. In turn, one expects $B^{\alpha\beta}$ to contain contributions that have a purely quantum mechanical origin. This aspect of $B^{\alpha\beta}$ is apparent in the total scattering that it is responsible for. From (2.6) and (2.11) one finds, for $\boldsymbol{k}$ constant and $\alpha \neq \beta$, that the total scattering is proportional to

$$\int_{-\infty}^{\infty} \mathrm{d}\omega\; B^{\alpha\beta}(\boldsymbol{k},\omega) = -\frac{1}{2N}\sum_a\sum_\gamma \varepsilon^{\alpha\beta\gamma}\langle S_a^\gamma\rangle + \frac{1}{N}\sum_{a,b}\sin\{\boldsymbol{k}\cdot(\boldsymbol{R}_a-\boldsymbol{R}_b)\}\langle S_a^\alpha S_b^\beta\rangle. \tag{2.16}$$

The first term in (2.16) is a direct consequence of the commutation relation for S_a^α and S_b^β, and it is evidently zero for a magnetic material with no long-range order. On using (2.8) one finds that the first term contributes to the cross-section for total scattering a factor

$$-(\hat{\boldsymbol{k}}\cdot\boldsymbol{P})(\hat{\boldsymbol{k}}\cdot\boldsymbol{M}) \tag{2.17}$$

where the magnetization is

$$\boldsymbol{M} = \frac{1}{N}\sum_a \langle \boldsymbol{S}_a\rangle.$$

Hence, the total scattering contains the projection of the magnetization on $\boldsymbol{k}$. 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 $\boldsymbol{k}$. Of course, $\boldsymbol{M}$ is zero for an antiferromagnet, and other configurations with complete compensation of the moments.

The second term in (2.16) is zero for a material in which the magnetic ions occupy sites that are centrosymmetric for, in this case, the two-spin correlation function depends only on $|\boldsymbol{R}_a-\boldsymbol{R}_b|$. 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 $B^{\alpha\beta}$ is most probably zero. Exceptions are provided by various novel materials, like a dimerized chain with anisotropic exchange interactions. Since the one-ion anisotropy Hamiltonian vanishes identically for spins of magnitude $\frac{1}{2}$, such as that for Cu^{2+}, the anisotropic interaction plays an important role in the magnetic anisotropy of antiferromagnetic cuprates and similar materials. Lastly, the second term in (2.16) contributes to the cross-section for total scattering a factor

$$(\hat{\boldsymbol{k}}\cdot\boldsymbol{P})(1/N)\sum_{a,b}\sin\{\boldsymbol{k}\cdot(\boldsymbol{R}_a-\boldsymbol{R}_b)\}\hat{\boldsymbol{k}}\cdot\langle\boldsymbol{S}_a\times\boldsymbol{S}_b\rangle. \tag{2.18}$$

It is interesting to observe that this contains the projection on $\boldsymbol{k}$ of a chiral order parameter [12].

We now discuss in yet more detail the physical properties of the symmetric, $A^{\alpha\beta}$, and antisymmetric, $B^{\alpha\beta}$, components of the van Hove response function. Prior to this, we notice that $B^{\alpha\beta}$ determines not only the polarization-dependent contribution to the cross-section but also the polarization created in the scattering of an initially unpolarized beam.

3. The response function

Initially, we look at properties of $S^{\alpha\beta}(\boldsymbol{k},\omega)$ that do not depend explicitly on the magnetic state of the target sample. The essential feature of the atomic variables in the interaction operator is that they are represented by Hermitian operators, e.g. $\{S_a^\alpha\}^+ = S_a^\alpha$.

First, on using the identity

$$\{S^{\alpha\beta}(\boldsymbol{k},\omega)\}^* = S^{\beta\alpha}(\boldsymbol{k},\omega) \tag{3.1}$$

it is at once evident that both $A^{\alpha\beta}$ and $B^{\alpha\beta}$ are purely real. This property, in turn, means that the cross-section is purely real, as it must be. Also,

$$A^{\alpha\beta}(\boldsymbol{k},\omega) = \mathrm{Re}\, S^{\alpha\beta}(\boldsymbol{k},\omega) \tag{3.2}$$

and

$$B^{\alpha\beta}(\boldsymbol{k},\omega) = -\mathrm{Im}\, S^{\alpha\beta}(\boldsymbol{k},\omega). \tag{3.3}$$

These results follow immediately from the definitions (2.10) and (2.11) and the identity (3.1).

Next, we use the identity which is often referred to as the condition of detailed balance, namely,

$$S^{\beta\alpha}(\boldsymbol{k},\omega) = \exp(\omega/T) S^{\alpha\beta}(-\boldsymbol{k},-\omega) \tag{3.4}$$

where T is the temperature in units of k_B. One finds

$$A^{\alpha\beta}(\boldsymbol{k},\omega) = \exp(\omega/T) A^{\alpha\beta}(-\boldsymbol{k},-\omega) \tag{3.5}$$

and

$$B^{\alpha\beta}(\boldsymbol{k},\omega) = -\exp(\omega/T) B^{\alpha\beta}(-\boldsymbol{k},-\omega). \tag{3.6}$$

Introducing functions $\Phi^{\alpha\beta}$ and $\Psi^{\alpha\beta}$ through the relations

$$A^{\alpha\beta}(\boldsymbol{k},\omega) = \{1+n(\omega)\}\Phi^{\alpha\beta}(\boldsymbol{k},\omega) \tag{3.7}$$

and,

$$B^{\alpha\beta}(\boldsymbol{k},\omega) = \{1+n(\omega)\}\Psi^{\alpha\beta}(\boldsymbol{k},\omega) \tag{3.8}$$

with, as usual, the so-called detailed-balance factor defined as

$$\{1+n(\omega)\} = \{1-\exp(-\omega/T)\}^{-1} \tag{3.9}$$

the results (3.5) and (3.6) lead to

$$\Phi^{\alpha\beta}(\boldsymbol{k},\omega) = -\Phi^{\alpha\beta}(-\boldsymbol{k},-\omega) \tag{3.10}$$

and

$$\Psi^{\alpha\beta}(\boldsymbol{k},\omega) = \Psi^{\alpha\beta}(-\boldsymbol{k},-\omega). \tag{3.11}$$

Results (3.10) and (3.11) are well known, and $\Phi^{\alpha\beta}(\boldsymbol{k},\omega)$ is frequently expressed in terms of an auxiliary function, e.g. the dissipative part of the susceptibility or Kubo's relaxation function [8, 9]. The result in [6] for the susceptibility, numbered (A7), has an incorrect energy denominator which affects all results that ensue.

Evaluated for $\omega = 0$, the result (3.6) shows that

$$B^{\alpha\beta}(\boldsymbol{k}, 0) + B^{\alpha\beta}(-\boldsymbol{k}, 0) = 0. \tag{3.12}$$

Thus, if the response function is independent of the sign of $\boldsymbol{k}$, and so the response is spatially isotropic, the conclusion from (3.12) is that the antisymmetric part of it is zero for $\omega = 0$. Also, in this case $\Psi^{\alpha\beta}$ becomes an even function of ω, and $\Phi^{\alpha\beta}$ an odd function.

The result (3.12) applies to the special case of Bragg scattering. A helical ordering of the atomic magnetic moments is an example of a non-collinear configuration of the moments for which $B^{\alpha\beta}(\boldsymbol{k}, 0)$ is an odd function of $\boldsymbol{k}$. In consequence, for a simple helix with a pitch $2\pi/Q$ the intensities of Bragg reflections are different for the satellites at the settings $\pm\boldsymbol{Q}$.

An additional set of useful results follow from the invariance of the Hamiltonian to a reversal of the velocities and rotations, or spin, of the electrons in the target sample. The time-reversal invariance of the Hamiltonian leads to the identity [9, 10]

$$S^{\alpha\beta}_{-\boldsymbol{H}}(-\boldsymbol{k}, \omega) = S^{\beta\alpha}_{\boldsymbol{H}}(\boldsymbol{k}, \omega) \tag{3.13}$$

where $\boldsymbol{H}$ is the applied magnetic field or the axis of quantization. All previous identities and results are valid for an arbitrary $\boldsymbol{H}$. For brevity of notation we have chosen not to attach a subscript $\boldsymbol{H}$ to the response function and quantities derived from it. Note that, just as in (3.4), the identity (3.13) entails different signs for $\boldsymbol{k}$, and this requirement is absent in the work reported in [6]. If the ions occupy sites that are centrosymmetric and there is no long-range order, or the magnetic order is collinear, the van Hove response function does not depend on the sign of $\boldsymbol{k}$.

Applied to $B^{\alpha\beta}_{\boldsymbol{H}}$, for which, by definition, $B^{\alpha\alpha}_{\boldsymbol{H}} = 0$, equation (3.13) leads to the revealing results

$$B^{\alpha\beta}_{\boldsymbol{H}}(\boldsymbol{k}, \omega) = \frac{\mathrm{i}}{2}\{S^{\alpha\beta}_{\boldsymbol{H}}(\boldsymbol{k}, \omega) - S^{\alpha\beta}_{-\boldsymbol{H}}(-\boldsymbol{k}, \omega)\} \tag{3.14}$$

and

$$B^{\alpha\beta}_{\boldsymbol{H}}(\boldsymbol{k}, \omega) + B^{\alpha\beta}_{-\boldsymbol{H}}(-\boldsymbol{k}, \omega) = 0. \tag{3.15}$$

The last result shows that $B^{\alpha\beta}_{\boldsymbol{H}}$ is possibly different from zero even in the absence of a preferred magnetic axis in the target sample, given that the ions occupy non-centrosymmetric sites and $\boldsymbol{k} \neq 0$. 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. $S^{xy}_{\boldsymbol{H}}(\boldsymbol{k}, \omega) \neq 0$ for $\boldsymbol{H} = 0$ and no spontaneous ordering of the moments.

Returning to (3.14), we can view $B^{\alpha\beta}_{\boldsymbol{H}}$ as a measure of the chiral signature of excitations. Consider, for example, $\boldsymbol{H}$ aligned with the z-axis. In this instance, $S^{xy}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$ describes excitations with circular polarization, the corresponding helicity is parallel to the z-axis, and $B^{xy}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$ is a difference between the left- and right-handed helicity states.

A simple, exact, and well-known example is the excitation of one spin-wave in a ferromagnet. One finds, for $\boldsymbol{H}$ along the z-axis and $\boldsymbol{P} = (0, 0, P_z)$,

$$B^{xy}_{\boldsymbol{H}}(\boldsymbol{k}, \omega) = \mathrm{i}S^{xy}_{\boldsymbol{H}}(\boldsymbol{k}, \omega) \tag{3.16}$$

and

$$\sum_{\alpha,\beta} G^{\alpha\beta}(\boldsymbol{P}, \boldsymbol{k}) B^{\alpha\beta}_{\boldsymbol{H}}(\boldsymbol{k}, \omega) = P_z \hat{k}_z^2 S\{n_{\boldsymbol{k}}\delta(\omega + \omega_{\boldsymbol{k}}) - (1 + n_{\boldsymbol{k}})\delta(\omega - \omega_{\boldsymbol{k}})\}. \tag{3.17}$$

Here, $\omega_{\boldsymbol{k}}$ is the dispersion of the spin-wave and $n_{\boldsymbol{k}} = n(\omega_{\boldsymbol{k}})$. We note that the explicit result is consistent with the general structure of the cross-section as it is exposed in (2.15).

From (3.17) one sees that created and annihilated spin-waves can be said to have equal and opposite helicities. We have previously noted the analogue of this effect in (elastic) Bragg diffraction from a magnet with a helical order of the moments. Adding to (3.17) the contribution made by $A^{\alpha\beta}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$ one finds that the creation and annihilation events in the cross-section are accompanied by the factors

$$(1 + \hat{k}_z^2 \pm 2P_z\hat{k}_z^2) \tag{3.18}$$

where the $-$ $(+)$ sign goes with creation (annihilation). If $\boldsymbol{k}$ is aligned with the preferred axis, i.e. $\hat{k}_z^2 = 1$, there is what amounts to a selection rule which operates for the ideal situations where $P_z = \pm 1$. These results were verified in experiments reported three decades ago [3].

As a second example, let us consider linear spin-waves in a ferromagnetically ordered material with a hexagonal close-packed (h.c.p.) structure. We can include single-ion anisotropy to make the model suitable for terbium metal, for example. The essential differences between the model and the simple ferromagnet considered in the preceding paragraphs is that the magnetic ions are not at centres of inversion symmetry, and, in addition, the anisotropy means that the total spin along the preferred axis is not a constant of motion. Even though the ions are not at centres of inversion symmetry $S^{xy}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$ does not depend on the sign of $\boldsymbol{k}$, and the same lattice structure factor occurs in both $A^{\alpha\beta}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$ and $B^{xy}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$. In consequence, the cross-section for linear spin-waves in the h.c.p. ferromagnet is proportional to (3.18) multiplied by the lattice structure factor.

A key aspect of the value found for $B^{xy}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$ is that it is largely determined by $[S^x, S^y]$. In the linear spin-wave approximation this commutator is taken to be a classical number, namely iS, not the operator iS^z. Kinematic interactions not included in the linear theory will thus modify the value found for $B^{xy}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$.

We do not have an exact result for a simple antiferromagnet. The standard spin-wave approximation, constructed from the Néel state, shows that the response function in (3.16) is zero. This result reflects the fact that the spin excitations on the two sublattices are the same except for the precession which is in opposite directions. The situation is changed by applying a magnetic field, because the field creates an imbalance in the two precession frequencies, and the quantity in (3.16) is different from zero. A calculation which confirms this argument is reported in [11] which examines the excitations in a magnetic salt with hybridized lattice and spin-wave modes.

The three examples in the preceding discussion have the common feature of a collinear configuration of the magnetic moments. For this case and linear spin excitation, $B^{\alpha\beta}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$ has for its dependence on ω the function shown in (3.17), and the form of this function reflects the fact that the commutator of the components of the spin operator perpendicular to the preferred axis is a c-number, i.e. the excitations are non-interacting bosons. As might be anticipated, linear excitations from a non-collinear configuration of the moments lead to a different kind of $B^{\alpha\beta}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$. While the dependence on ω is derived from the function in (3.17), because the spin excitations do not interact, one finds $B^{\alpha\beta}_{\boldsymbol{H}}(\boldsymbol{k}, \omega) \neq B^{\alpha\beta}_{\boldsymbol{H}}(-\boldsymbol{k}, \omega)$. As our example, we consider a simple helix with a pitch $2\pi/Q$. Here, there are spin-wave excitations about the two satellite settings $\pm\boldsymbol{Q}$, each with the ω-dependence given in (3.17). However, the structure factors at $\pm\boldsymbol{Q}$, not surprisingly, are different and such that $B^{\alpha\beta}_{\boldsymbol{H}}(\boldsymbol{k}, \omega)$ is an odd function of $\boldsymbol{k}$. The latter finding is for inelastic scattering the analogue of the result noted before about Bragg intensities in diffraction by a helimagnet.

4. Conclusions

We have shown that the contribution to the cross-section for the magnetic scattering of neutrons which is induced by polarization in the primary beam is the product of two factors: (i) the projection of the polarization, $\boldsymbol{P}$, on the scattering vector, $\boldsymbol{k}$, and (ii) the projection of the antisymmetric combination of the Cartesian components of the van Hove spin response function on $\boldsymbol{k}$ (the contribution in question is not included in the discussion reported in [13]). The antisymmetric combination we denote by $B_{\boldsymbol{H}}^{\alpha\beta}(\boldsymbol{k},\omega) = -B_{\boldsymbol{H}}^{\beta\alpha}(\boldsymbol{k},\omega)$, where ω is the energy transferred to the sample in the scattering event and $\boldsymbol{H}$ is the preferred magnetic axis. The cross-section in question is proportional to

$$(\boldsymbol{k}\cdot\boldsymbol{P})\sum_{\alpha\beta\gamma}\varepsilon^{\alpha\beta\gamma}k_\gamma B_{\boldsymbol{H}}^{\alpha\beta}(\boldsymbol{k},\omega)$$

where $\varepsilon^{\alpha\beta\gamma}$ is the completely antisymmetric tensor with three Cartesian indices.

It is shown that $B_{\boldsymbol{H}}^{\alpha\beta}(\boldsymbol{k},\omega)$ is proportional to the imaginary part of the van Hove spin response function, and intimately connected to the commutation (Lie) algebra of spin operators. Two classes of properties are derived for $B_{\boldsymbol{H}}^{\alpha\beta}(\boldsymbol{k},\omega)$ by applying to the response function, respectively, the condition of detailed balance, and the invariance of the target sample's properties to a reversal of the direction of time.

For elastic scattering $\omega = 0$, and one finds

$$B_{\boldsymbol{H}}^{\alpha\beta}(\boldsymbol{k},0) + B_{\boldsymbol{H}}^{\alpha\beta}(-\boldsymbol{k},0) = 0$$

with $B_{\boldsymbol{H}}^{\alpha\beta}(\boldsymbol{k},0) = B_{-\boldsymbol{H}}^{\alpha\beta}(\boldsymbol{k},0)$. The spin response function can be spatially anisotropic, and different for the scattering wave vectors $\boldsymbol{k}$ and $-\boldsymbol{k}$, when the lattice sites occupied by the spins are not centrosymmetric, and the configuration adopted by the spin moments is not collinear.

One can view $B_{\boldsymbol{H}}^{\alpha\beta}(\boldsymbol{k},\omega)$ as a measure of the chiral signature of spin excitations [12]. This property is very clear in the total scattering, for the corresponding cross-section contains the projection on $\boldsymbol{k}$ of a chiral order parameter. In inelastic scattering events, $B_{\boldsymbol{H}}^{\alpha\beta}(\boldsymbol{k},\omega)$ is properly interpreted as the difference in the response of excitations with left- and right-handed helicities. Several examples of inelastic scattering are discussed, which illustrate the roles of the spin algebra and the configuration of the equilibrium distribution of the spin moments.

The cross-section for the magnetic scattering of unpolarized neutrons, $\boldsymbol{P} = \boldsymbol{0}$, is expressed in terms of the symmetric combination of the Cartesian components of the van Hove spin response function, which is the real part of the response function. We compare and contrast the properties of the symmetric and antisymmetric combinations, using the condition of detailed balance and time-reversal invariance.

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