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Abstract. Studies of diffuse magnetic scattering largely benefit from the use of a multi-detector covering wide scattering angles. Therefore, the different contributions to the diffuse scattering that originate from magnetic, nuclear coherent, and nuclear spin-incoherent scattering can be separated by the so-called XYZ-polarization analysis. In the past this method has been successfully applied to the analysis of diffuse scattering by polycrystalline samples of magnetic disordered materials. Single crystal studies that exploit the vector properties of spin correlations are of particular interest for furthering our understanding of frustration effects in magnetism. Based on the symmetry properties of polarised scattering a suitable extension of the conventional XYZ method has been derived, which allows for the complete separation and the analysis of features of diffuse magnetic scattering from single crystals.

1. Introduction

The scattering theory of polarised neutrons including magnetic interactions started with the early work of by Halpern and Johnson [1] and has been essentially completed by Blume[2] and Maleyev[3] more than fourty years ago. The major milestones in development of experimental means and applications have been set by technique of longitudinal polarisation analysis (Moon *et al.* [4]) and by spherical neutron polarimetry (Tasset and Brown) [5, 6], which nowadays provides a high precision tool to analyse the full polarisation tensor for single crystal magnetic scattering. The development for diffuse scattering utilizing a wide angle detector has been essentially driven by Schärpf, who established with *his* D7 instrument at the ILL and the so-called XYZ method[7, 8] efficient means for measuring and analysing spin-correlations in powder samples. With recent instrumental progress, the D7 at ILL [9] and the DNS at JCNS [10] have gained the required efficiency to explore more routinely the diffuse magnetic scattering from single crystals. To overcome the pending problem that the XYZ-method is invalid for the separation of the more complex scattering from single crystals, here an appropriate extension of the XYZ-method is derived using the symmetry properties of the polarised scattering.

2. Polarised neutron scattering and symmetry

According to Blume [2] and Maleyev [3] the neutron scattering process including magnetic interactions can be completely described by two master equations, Eq.(1) for the scattering cross-section, here for brevity denoted by the intensity I, and Eq.(2) for $\mathbf{P}'I$, where $\mathbf{P}'$ denotes the final polarization:

$$I = N^{\dagger}N + I_{si} + \mathbf{M}_{\perp}^{\dagger}\mathbf{M}_{\perp} + \mathbf{P}\cdot\mathbf{M}_{\perp}^{\dagger}N + \mathbf{P}\cdot\mathbf{M}_{\perp}N^{\dagger} + \mathrm{i}\mathbf{P}(\mathbf{M}_{\perp}^{\dagger}\times\mathbf{M}_{\perp}) \qquad (1)$$

$$\begin{aligned} \mathbf{P}'I &= \mathbf{P}(N^{\dagger}N - \frac{1}{3}I_{si}) + (\mathbf{P} \cdot \mathbf{M}_{\perp}^{\dagger})\mathbf{M}_{\perp} + (\mathbf{P} \cdot \mathbf{M}_{\perp})\mathbf{M}_{\perp}^{\dagger} - \mathbf{P}(\mathbf{M}_{\perp}^{\dagger}\mathbf{M}_{\perp}) \\ &\quad + \mathrm{i}N(\mathbf{P} \times \mathbf{M}_{\perp}^{\dagger}) - \mathrm{i}N^{\dagger}(\mathbf{P} \times \mathbf{M}_{\perp}) + N\mathbf{M}_{\perp}^{\dagger} + N^{\dagger}\mathbf{M}_{\perp} - \mathrm{i}(\mathbf{M}_{\perp}^{\dagger} \times \mathbf{M}_{\perp}) \,, \end{aligned} \tag{2}$$

where $N(\mathbf{Q}) = \sum_n b_n \exp(\mathrm{i}\mathbf{Q} \cdot \mathbf{R_n})$ is the nuclear structure factor and $\mathbf{M}_{\perp} = \mathbf{e_Q} \times \mathbf{M}(\mathbf{Q}) \times \mathbf{e_Q}$, is the vector of dipolar magnetic interaction of magnetic moments with the neutron spin, and $\mathbf{e_Q}$ is the unit vector along the direction of the scattering vector $\mathbf{Q}$. Accordingly, the parallel components to the scattering vector $\mathbf{Q}$ do not contribute to magnetic scattering. Here, $\mathbf{M}(\mathbf{Q}) = \sum_n \mathbf{M}_n \exp(\mathrm{i}\mathbf{Q} \cdot \mathbf{R_n})$ denotes the Fourier transform of the magnetic moments. I_{si} denotes the nuclear spin-incoherent scattering, assuming that the nuclear spins are randomly oriented. This diffuse background, which is usually small compared to magnetic and nuclear Bragg peaks, can be relatively large when considering diffuse magnetic and diffuse nuclear scattering.

The full information about the scattering terms in Eqs. (1,2) can be retrieved by spherical neutron polarimetry [6]. Therefore, the most convenient and standard choice of a x,y,z-coordinate system for the setting of the polarisation $\mathbf{P}$ is to have one axis x parallel to $\mathbf{Q}$, with the axes y and z perpendicular to $\mathbf{Q}$ pointing in- and out-of the scattering plane respectively.

However, when using multi-detectors $\mathbf{P}$ can be set ideally parallel to $\mathbf{Q}$ only for a single detector. Therefore, the XYZ-polarisation analysis for multi-detectors the axes x and y are chosen to be in the (horizontal) scattering plane and z to point (vertically) out of the scattering plane. In an excellent contribution to the present subject, the polarised scattering has been revisited and treated within the density matrix formalism by Schärpf [11]. Following Ref.[11] we consider first the general case of an arbitrarily rotated x,y,z-system and the polarised intensities that can be measured in spin-flip and non-spin flip modes and their relation to the above equations. With the definition of polarised intensities and polarisation

$$I = I_{\nu\nu} + I_{\nu\bar{\nu}} \quad , \quad P'_{\nu}I = I_{\nu\nu} - I_{\nu\bar{\nu}} \tag{3}$$

for any cartesian coordinate $\nu = x, y, z$, the non-spin flip (*nsf*) and spin-flip intensities (*sf*), $I_{\nu\nu}$ and $I_{\nu\bar{\nu}}$ respectively, have been derived [11] from Eqs. (1) to (3):

$$\begin{aligned} I_{\nu\nu} &= N^{\dagger}N + NM_{\perp\nu}^{\dagger} + N^{\dagger}M_{\perp\nu} + M_{\perp\nu}^{\dagger}M_{\perp\nu} + \frac{1}{3}I_{si} \\ I_{\nu\bar{\nu}} &= \mathbf{M}_{\perp}^{\dagger}\mathbf{M}_{\perp} - M_{\perp\nu}^{\dagger}M_{\perp\nu} + \mathrm{i}(\mathbf{M}_{\perp}^{\dagger} \times \mathbf{M}_{\perp})_{\nu} + \frac{2}{3}I_{si} \,. \end{aligned} \tag{4}$$

Eq. 4 shows a well known result namely that only components of $\mathbf{M}_{\perp}$ parallel to the neutron spin appear in the non-spin flip scattering and only components of $\mathbf{M}_{\perp}$ perpendicular to the neutron spin can contribute to the spin-flip scattering and is exemplified for $\nu = x$

$$I_{x\bar{x}} = M_{\perp y}^{\dagger}M_{\perp y} + M_{\perp z}^{\dagger}M_{\perp z} + \mathrm{i}(M_{\perp y}^{\dagger}M_{\perp z} - M_{\perp z}^{\dagger}M_{\perp y}) + \frac{2}{3}I_{si} \,.$$

It is further worthwhile to note that with any specific choice of the coordinate system the average of non-spin flip and spin-flip intensities differ for the different directions

$$I_x = \frac{1}{2}(I_{xx} + I_{x\bar{x}}) \neq I_y \neq I_z.$$

So far we followed the analysis of Schärpf. For a further analysis and separation of terms in the polarised neutron scattering, here, we continue with separating intensities in symmetric and antisymmetric parts. Therefore, we consider scattering for reversed polarisation by the

intensities $I_{\bar{\nu}\bar{\nu}}$ and $I_{\bar{\nu}\nu}$ which are experimentally likewise accessible, and we obtain the analogue to Eq. (4) from Eqs. (1-3):

$$
\begin{aligned}
I_{\bar{\nu}\bar{\nu}} &= N^\dagger N - N M_{\perp\nu}^\dagger - N^\dagger M_{\perp\nu} + M_{\perp\nu}^\dagger M_{\perp\nu} + \frac{1}{3} I_{si} \\
I_{\bar{\nu}\nu} &= \mathbf{M}_\perp^\dagger \mathbf{M}_\perp - M_{\perp\nu}^\dagger M_{\perp\nu} - \mathrm{i}(\mathbf{M}_\perp^\dagger \times \mathbf{M}_\perp)_\nu + \frac{2}{3} I_{si} \; .
\end{aligned}
\tag{5}
$$

In comparison to Eq. 4 and 5, a further simplification is achieved by considering the average non-spin-flip and spin flip intensities, Σ_ν^{nsf} and Σ_ν^{sf} respectively:

$$
\begin{aligned}
\Sigma_\nu^{nsf} = \frac{1}{2}(I_{\nu\nu} + I_{\bar{\nu}\bar{\nu}}) &= N^\dagger N + M_{\perp\nu}^\dagger M_{\perp\nu} + \frac{1}{3} I_{si} \\
\Sigma_\nu^{sf} = \frac{1}{2}(I_{\nu\bar{\nu}} + I_{\bar{\nu}\nu}) &= \mathbf{M}_\perp^\dagger \mathbf{M}_\perp - M_{\perp\nu}^\dagger M_{\perp\nu} + \frac{2}{3} I_{si}
\end{aligned}
\tag{6}
$$

showing the expected property of the unpolarised intensity I

$$
I = \Sigma_x = \Sigma_y = \Sigma_z = \Sigma_\nu^{nsf} + \Sigma_\nu^{sf} = N^\dagger N + \mathbf{M}_\perp^\dagger \mathbf{M}_\perp + I_{si} \; .
$$

By this we have eliminated the interference terms related to nuclear-magnetic correlations (spin-orbit coupling) and vector chirality terms (cross products) that now appear in the corresponding averaged deviations Δ_ν^{nsf} and Δ_ν^{sf} respectively, :

$$
\begin{aligned}
\Delta_\nu^{nsf} = \frac{1}{2}(I_{\nu\nu} - I_{\bar{\nu}\bar{\nu}}) &= N M_{\perp\nu}^\dagger + N^\dagger M_{\perp\nu} = 2\Re(N M_{\perp\nu}^\dagger) \\
\Delta_\nu^{sf} = \frac{1}{2}(I_{\nu\bar{\nu}} - I_{\bar{\nu}\nu}) &= 2\mathrm{i}(\mathbf{M}_\perp^\dagger \times \mathbf{M}_\perp)_\nu \; .
\end{aligned}
\tag{7}
$$

Eq. (7) reveals a well known result that is the basis of experiments with polarized neutrons using polarisation reversal without polarisation analysis. For the purpose of separating the interference terms due to chirality or due to nuclear-magnetic correlations, polarisation analysis is actually not required and can be achieved by setting ν, with polarisation reversal, either parallel (for chirality) or perpendicular (for nuclear-magnetic interference) to the scattering vector $\mathbf{Q}$. However, as can be seen, Eq. (7) also provides a complete separation of these terms for any choice of the cartesian coordinate system if polarisation analysis is applied.

Finally, one may note that it is also straightforward to derive this symmetry decomposition from Brown's tensor representation of polarised scattering [6] for the case of the standard coordinate system ($x\|\mathbf{Q}$) and also to include spin-incoherent scattering.

3. XYZ-polarisation analysis, powders and single crystals

The standard XYZ-method has been successfully applied in many studies of magnetic correlations using powder samples. From the directional dependence of the polarised scattering, the purely magnetic scattering contribution can be separated by experimental means of polarisation analysis. For further interest the reader is referred to a recent and detailed review [9] and a recent own study [12]. Here we shall briefly recall the fundamental equations (Eq. 8), and discuss the additional potential of instruments like DNS and D7 for studies of powder samples with polarisation analysis. An interesting consequence is that the interference of nuclear and induced magnetic scattering could be observed even for powders if chiral domains relate to satellites with different Q moduli or in presence of a symmetry breaking external field, whereas usually, chiral scattering is found in single crystals with a preferred domain orientation. In the following (3.2) the central issue will be to separate and analyse the different contributions to the polarised scattering from single crystals based on the conventional tools of the standard XYZ method. Hence, the separation will be achieved without approximations only from diagonal intensity elements.

3.1. The application to powders

In case that orientational averaging applies, e. g. for powder samples, we do not distinguish the components $M_{\perp\nu}$, and the magnetic scattering can be separated via the XYZ polarisation analysis [7, 8] from the diagonal intensities. In case of only weak guide fields one does not need to distinguish the intensities with respect to the sign of polarisation as proposed in the previous section.   It is a particular virtue of the XYZ-method that this method applies even if the Cartesian coordination system is arbitrarily rotated in the scattering plane, so that the new coordinates are x', y', z (see figure). In case of multi-detectors covering a larger Q-range it is trivially impossible to have the polarisation simultaneously parallel to all different Q vectors; the difference between the polarisation axis x' and the actual **Q** vector is the *Schärpf angle* α.
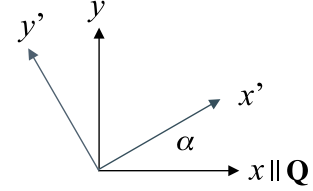
The information about the in-plane magnetic intensities, the square of magnetic amplitudes $\mathbf{M}_{\perp y}^2$, is recovered from $\mathbf{M}_{\perp y}^2 = \mathbf{M}_{\perp x'}^2 + \mathbf{M}_{\perp y'}^2 = \mathbf{M}_{\perp y}^2(\sin^2\alpha + \cos^2\alpha)$. Hence, the magnetic, nuclear coherent and spin-incoherent scattering can be separated by combinations of polarised intensities [7, 8, 9]:

$$
\begin{aligned}
|\mathbf{M}_\perp|^2 &= 2(I_{x'\bar{x}'} + I_{y'\bar{y}'} - 2I_{z\bar{z}})^{sf} = -2(I_{x'x'} + I_{y'y'} - 2I_{zz})^{nsf} \qquad (8)\\
I_{si} &= \frac{3}{2}\left(-I_{x'\bar{x}'} - I_{y'\bar{y}'} + 3I_{z\bar{z}}\right)\\
N^2 &= I_{zz} - \frac{1}{2}|\mathbf{M}_\perp|^2 - \frac{1}{3}I_{si}.
\end{aligned}
$$

Here, we have assumed that the intensities do not change for a common polarisation reversal of initial and final polarisation. More carefully, for the application of this analysis we have to consider whether the antisymmetric part actually vanishes. This is fulfilled only if there is no distinguished direction by any external field. Guide fields applied in the XZY method are typically in the order of magnitude of ten Gauss and they are too weak to induce any significant sample magnetization in paramagnetic or antiferromagnetic samples.

Strong fields invalidate the standard XYZ separation method, however, this allows for the study of the correlations and interference terms between nuclear and magnetic scattering amplitudes by determining the antisymmetric part of the polarised intensity, *id est* measuring intensity differences for polarisation reversal. A unique response of chiral terms to an applied horizontal external field is less likely, although not impossible. A well-known strategy for single crystals used in a "half-polarised" setup [13] can also be applied to powder diffraction. Such a determination of correlations between nuclear and magnetic scattering amplitudes from powders looks promising particularly for detectors covering a large solid angle without polarisation analysis. With respect to polarisation analysis of scattering it may be advantageous that only the difference of the *nsf*-intensities contributes to the signal, and the background from *sf*-scattering could be eliminated.

3.2. The application to single crystals

For a single crystal, even without applied external field, its intrinsic anisotropy and possible polarity may give rise to antisymmetric scattering contributions. Therefore, the symmetry decomposition resulting in Eqs. (6,7) is important and helpful to analyse the polarised scattering and its magnetic contributions. For a Cartesian coordinate system (x', y', z) rotated by the angle α around the vertical z-axis with respect to x parallel to **Q**, the following relations can be derived from Eqs. (6) and (7) providing a complete separation

$$N^\dagger N \;=\; \frac{1}{2}\left(\Sigma_x^{nsf} + \Sigma_y^{nsf} - \Sigma_z^{sf}\right) = \frac{1}{2}(\Sigma_{x'}^{nsf} + \Sigma_{y'}^{nsf} - \Sigma_z^{sf}) \tag{9}$$

$$I_{si} \;=\; \frac{3}{2}(\Sigma_x^{nsf} - \Sigma_y^{nsf} + \Sigma_z^{sf}) = \frac{3}{2}\frac{\Sigma_{x'}^{nsf} - \Sigma_{y'}^{nsf}}{\cos^2\alpha - \sin^2\alpha} + \frac{3}{2}\Sigma_z^{sf} \tag{10}$$

$$M_{\perp y}^\dagger M_{\perp y} \;=\; \Sigma_z^{sf} - \frac{2}{3}I_{si} \tag{11}$$

$$M_{\perp z}^\dagger M_{\perp z} \;=\; \Sigma_z^{nsf} - \frac{1}{3}I_{si} - N^\dagger N \tag{12}$$

$$I_{chiral,x'} \;=\; 2i(\mathbf{M}_\perp^\dagger \times \mathbf{M}_\perp)_{x'} = \Delta_{x'}^{sf} \;\; = \cos\alpha\,\Delta_x^{sf} \tag{13}$$

$$I_{NM,y'} \;=\; 2\Re(N Q_{y'}^\dagger) \quad\;\;\; = \Delta_{y'}^{nsf} = \cos\alpha\,\Delta_y^{nsf} \tag{14}$$

$$I_{NM,z'} \;=\; 2\Re(N Q_{z'}^\dagger) \quad\;\;\; = \Delta_{z'}^{nsf} = \quad\;\; \Delta_z^{nsf}\;, \tag{15}$$

where the geometrical correction factors in Eqs. (10,13,14) stem from the projections $M_{\perp x'} = \sin\alpha\, M_{\perp y}$ and $M_{\perp y'} = \cos\alpha\, M_{\perp y}$ to the scattering vector[1]. Compared to the information content of powder experiments, the study of diffuse polarised scattering from single crystals yields additionally the anisotropy of magnetic correlation functions, possible vector chirality and all nuclear- magnetic correlations, essential information about complex magnetic materials of high current interest. It is worthwhile to note that a separation of the chiral term has already been given earlier by Eq. 50 in Ref.[8].

For diffuse scattering the typical experimental strategy is to map out scattering planes by stepwise rotations of the crystal around the vertical axis z. Note, the separation by Eqs. (9) to (15) is independent of the sample rotation ω and the convenient choice is a fixed coordinate system x', y', z.

In order to interpret the magnetic scattering amplitudes in terms of the crystal lattice, the in-plane component $M_{\perp y}$ has to be decomposed further into independent lattice components in the plane (ab) and is given by the projections of M_a and M_b, while the vertical component is simply identified by M_c. For orthogonal components, only the squares of M_a and M_b determine $M_{\perp y}$, see Fig. 1. Note, this example demonstrates that the considerations here also apply to inelastic scattering. The disentanglement of M_a and M_b is, analog to structure determination
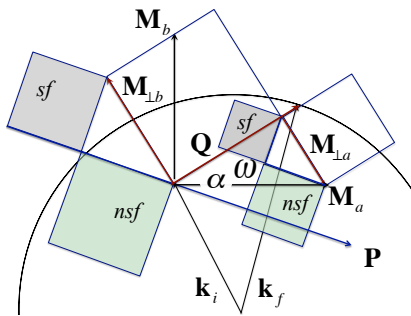
Figure 1. (color online) Polarisation $\mathbf{P}$ and magnetic (inelastic) scattering $M_{\perp\nu}^2$ (white squares) for the case of a *Schärpf angle* α between scattering vector $\mathbf{Q}$ and polarisation $\mathbf{P}$; here ω denotes the angle between $\mathbf{Q}$ and reciprocal lattice vector $\mathbf{a}^*$ with $\mathbf{M}_a\|\mathbf{a}^*$. Shaded squares represent *sf*- and *nsf*-intensities related to the projections of $\mathbf{M}_{\perp a}$ and $\mathbf{M}_{\perp b}$ $\perp$ and $\|\mathbf{P}$ respectively. The vertical component ($\|z$ and $\perp$ to $\mathbf{P}$, *sf*-intensity) is not shown.

with short-range order and displacements, relying on sufficient information in extended Brillouin

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[1] In view of the singularity at $\alpha = \pi/4$ in Eq.(10) and for smallest possible corrections, the polarisation axis x' is best set parallel to the $\mathbf{Q}$ vector for the central part of a multidetector. For a multidetector accepting scattering within angles $\Delta\vartheta = 120^o$, the *Schärpf angles* α (between $\mathbf{Q}$ and $\mathbf{P}_x$) will be equal to the deviations from the average Bragg angle $-30^o \leq \alpha < 30^o$; hence, correction factors vary between 1 and 0.5 for the spin-incoherent scattering, between 1 and $\sqrt{3}/2$ for the chiral contribution (Eq. 13) and likewise for the nuclear magnetic interference (Eq. 14) resulting in small average corrections $3\sqrt{3}\,/\,2\pi \approx 0.827$ and $\pi/3 \approx 0.955$ respectively.

zones of reciprocal space. Diffuse scattering related to disorder phenomena typically implies an exponential decay of correlations in real space. Hence a Fourier analysis will efficiently reveal finite real space pair correlation functions, an approach that is a linear least squares problem having a unique solution. Therefore, the determination of three dimensional pair-correlations functions requires accordingly measurements of more than a single scattering plane.

4. Final remarks

With the substantial gain in efficiency seen with the developments on instruments in the recent years, particularly on the DNS and D7, single crystal studies with polarisation analysis became feasible and produced already first outstanding results [14]; a further example can also be found here in the proceedings of the PNSXM 2009 [15]. For a systematic and thorough understanding the separation of the different components is essential, a task that to date has been believed to be incapable by the conventional means used in XYZ polarisation analysis. The solution here, utilizing the symmetry properties, also places the emphasis on additional measurements with polarisation reversal. It is worthwhile to note that this request does not demand for more beamtime to achieve equal statistical weights.

So far we have not considered the off-diagonal terms. These terms also represent the interference terms, Eqs. (13-15), due to chiral correlations and due to coupling of spin-space and real-space variables. Spherical neutron polarimetry of off-diagonal terms distinguishes these effects irrespectively of a possible depolarisation from different magnetic domains in the crystal. Obviously, the diagonal antisymmetric polarised intensities Eq. (7) also reveal the mentioned interference terms. It should be noted that the same XYZ-setup for multi-detectors can be used to access the complete polarisation tensor and off-diagonal terms can be determined with the same accuracy by spherical neutron polarimetry with precessing incident polarization[16, 17]. However, the here proposed XYZ-polarisation analysis, a method based on only diagonal terms and applicable for multi-detectors, will certainly provide a much more convenient and efficient separation for the diffuse magnetic neutron scattering from single crystals.

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