Analysis of Neutron Polarimetry data using MuFit

Amy Poole, Bertrand Roessli
Laboratory for Neutron Scattering, Paul Scherrer Institut, CH-5232 Villigen, PSI, Switzerland
E-mail: amy.poole@psi.ch, bertrand.roessli@psi.ch

Abstract. This paper presents a new single crystal neutron scattering data analysis program, MuFit. The program can be used to refine spherical neutron polarimetry (SNP) data as well as nuclear and magnetic single crystal diffraction data. The article presents a brief overview of the SNP process and the core mathematics used to calculate the SNP matrices and gives an example of data that has been fitted using the MuFit package, which is available on request from the author: Bertrand.Roessli@psi.ch.

1. Neutron Polarimetry
Spherical neutron polarimetry (SNP) is the measurement of the both the longitudinal and transverse components of a polarized neutron beam after the scattering event. It is the experimental realization of the independent, theoretical work of Blume and Maleev who put forward a general description of the scattering cross section for a neutron beam that includes an arbitrary initial and final polarization, [1, 2, 3]. Their work was an extension of that by Moon, Riste and Koehler who developed longitudinal polarization analysis (LPA) and a set of equations to describe the observed experimental intensities [4]. LPA type experiments require a guide field to maintain the polarization of the neutron beam. Any transverse components of the polarization vector precess around this field and the information contained in these components is lost. CRYOPAD (CRYOgenic Polarization Analysis Device) and, subsequently, MuPAD (MU-metal Polarization Analysis Device) prevent this information loss by shielding the polarized beam from stray magnetic fields, which negates the requirement for a guide field and allows SNP-type measurements [6, 7]. The power of the technique lies in the accuracy with which one may determine the orientation of $M_\perp(Q)$ and the ability to determine the change of intensity when the structure has out of phase components [8, 9, 10].

2. Experimental Process
The neutron polarimetry process may be considered in three key stages. The first step is to orient a polarized neutron beam, usually in one of the directions, $x$, $y$ or $z$ where $x$ is in the direction of the scattering vector $Q$, $z$ is along the instrument axis, perpendicular to the scattering plane and $y$ makes the right hand set [11]. The oriented, polarized beam then enters the ‘zero-field’ chamber where it meets the sample. On scattering, the neutron spins are rotated by the magnetic scattering vector, $M_\perp(Q)$, that lie in the direction of polarization and flipped by components that are perpendicular. The third step is then to measure the intensity of the spin-flip and non spin-flip scattering in each of the directions $x$, $y$ and $z$. In the case of purely magnetic scattering the process may be visualized as a rotation about $M_\perp(Q)$, but in which...
polarization may be created along, $Q$, when $M_{\perp}(Q)$ has both real and imaginary components, i.e. for helical and cycloidal structures [12].

3. Mathematical Summary

The spin-flip and non-spin flip scattering intensities, $I^-$ and $I^+$ respectively, are measured for each of the initial $X$, $Y$ and $Z$ and final $x$, $y$ and $z$ directions. The $3 \times 3$ polarization matrix,

$$
\begin{pmatrix}
P_{Xx} & P_{Xy} & P_{Xz} \\
P_{Yx} & P_{Yy} & P_{Yz} \\
P_{Zx} & P_{Zy} & P_{Zz}
\end{pmatrix},
$$

is found from these 18 measurements as each element of the matrix is given by,

$$
P_{If} = I^{+}_f - I^{-}_f
$$

The measured intensity, $I_{If}$, is a sum of the scattering from all of the $n$ magnetic domains, weighted by the volume fraction $\delta^n$, that are present in the sample:

$$
I_{If} = \sum_n \delta^n I^n_{If}
$$

To determine the intensity of the polarized scattering in the purely elastic case the general scattering cross section has the form,

$$
\left( \frac{d\sigma}{d\Omega} \right) = \sum_{\sigma_I\sigma_f} p_{\sigma_I} p_{\sigma_f} \times |\langle \sigma_f | \hat{V}(Q) | \sigma_I \rangle|^2,
$$

where $\sigma_I$ and $\sigma_f$ are spinors that describe the incident and final polarization respectively and $p_{\sigma_I}$ and $p_{\sigma_f}$ describe the efficiency of that polarization. The term $\hat{V}(Q)$ is the Fourier transform of the interaction potential between the incident neutron and the target system multiplied by $(m/2\pi\hbar)$, [13]. When the polarization of the beam is not unitary, both before and after the sample, a sum has to be made over all the combinations of $\sigma_I$ and $\sigma_f$ which are weighted according to the polarization efficiency. If we assume that there is no contribution to the magnetism from the nuclear spin the term $\hat{V}(Q)$ may be decomposed into parts that describe the nuclear interaction $\hat{V}_N(Q)$ and the magnetic interaction $\hat{V}_M(Q)$,

$$
\hat{V}(Q) = \hat{V}_N(Q) \cdot 1 + \hat{V}_M(Q) \cdot \sigma.
$$

The term $\hat{V}_N(Q)$ is multiplied by a $2 \times 2$ unitary matrix, represented by $1$, and $\hat{V}_M(Q)$ is multiplied by a vector of Pauli matrices, $\sigma$. In this form the potential can be considered as a rotational matrix that operates on the initial spin state $\sigma_I$ to give the final spin state $\sigma_f$. The measurement of the intensity of the neutrons with final spin states $\sigma_f^+$ and $\sigma_f^-$ reveals the rotational operation that has occurred and from this the potential may be deduced.

If we consider the final polarization vector $P'_f$, with initial polarization $P_I$, where $I = X$, $Y$ or $Z$ we have the polarization vector described in the Blume-Maleev equations:

$$
P'_I = (P'_{Ix}, P'_{Iy}, P'_{Iz}) = \frac{\text{Tr} \cdot \hat{\rho}|\hat{V}'\rangle\langle\hat{V}'|}{\text{Tr} \cdot |\hat{V}'|^2}
$$

where $\hat{V}'$ is the complex conjugate transpose of $\hat{V}$, $\text{Tr}$ denotes the trace over neutron spin variables and $\hat{\rho}$ is a probability density matrix, described by,

$$
2\hat{\rho} = 1 + P_I \cdot \hat{\sigma}.
$$
From the expansion of equation 5 it is possible to derive the Blume-Maleev equations, [1, 2, 3],

\[
P'_I = [(P_I^* N N^*) + (-P_I (M_{N} \cdot M_{N}^*) + (M_{N} (P_I \cdot M_{N}^*) + M_{N}^* (P_I \cdot M_{N}))
+ (-i(M_{N}^* \times M_{N}))
+ (N M_{N}^* + N^* M_{N} - i(N M_{N}^* - N^* M_{N} \times P_I)])/\sigma,
\]

\[
\sigma = (N N^*) + (M_{N} \cdot M_{N}^*)
+ (iP_I \cdot (M_{N}^* \times M_{N})) + (P_I \cdot (M_{N} N^* + M_{N}^* N)).
\]  

Where \( M_N \) is the magnetic interaction vector, \( M_N^* \) is the complex conjugate of the magnetic interaction vector and \( N \) is the nuclear structure factor. The \( Q \) dependence of \( M_N \) and \( N \) has been omitted for clarity. The first and second rows of equation 7 give the nuclear and magnetic intensities respectively. The third row describes the contribution to the scattering from the real and imaginary components of \( N \). The final row gives the interference effects from coincident nuclear and magnetic scattering and is zero in all cases where \( k \neq 0 \), [14].

4. Data Analysis

To analyze and fit the polarimetry data, dedicated software was developed in the interpreted language Octave, which is an open source software developed to perform numerical and mathematical operations [15]. The Octave language has the advantage that it is mostly compatible with Matlab and hence is easily accessible to most users. Furthermore, it has the option to dynamically load C++ functions to reduce long computation times.

Currently MuFit can be used to calculate the nuclear and magnetic structure factors including anisotropic thermal factors, and extinction corrections, however, absorption corrections cannot be applied. Furthermore, a simultaneous refinement of nuclear and magnetic structures cannot be performed. The magnetic structure factors are calculated by making use of the Fourier components of the atomic moment, analogous to the approach taken in FullProf [16]. The weighted population of the symmetry related twins and rotationally related magnetic domains is summed and the weighting may be refined, as described in equation 2 to give the correct final polarization. Structures that are described by more than one \( k \) vector cannot currently be calculated within the program. For comparison the Blume-Maleev equations have been programmed and can be used to calculate the matrices, the equations are not used to fit the data as the depolarization corrections and domain populations cannot be applied within the vector formalism adopted by the equations. To correct the polarization matrices for the finite polarization of the beam the MuFit software calculates the neutron cross sections weighted by the beam polarization, \( p_{RF} \) and \( p_{RF} \), and sums to find the final intensity in each channel, equation 3. The step described in equation 3 describes the ‘leakage’ of intensity from one polarization channel to another and is not explicitly included in the Blume-Maleev polarization equations.

In order to speed up the calculations, the mathematical operations were vectorized as much as possible and C++ was used for the calculation of the elements of the polarization matrix to avoid loops that are costly in terms of computing time as no Just-In-Time compiler is included in Octave. To perform least-square refinements with MuFit, the user has the choice between three algorithms [17]: Levenberg-Marquardt, Simplex and Simulated Annealing. These routines have been implemented to fit nuclear and magnetic intensities from single crystals and polarimetry data. The program has been used to determine the magnetic structure of ErMn\(_2\)O\(_5\) [18] and BaMnF\(_4\) [10] and were found to be in agreement with the CCSL crystallographic library [19]. A plot of the goodness of fit of the nuclear and magnetic calculations for BaMnF\(_4\) is shown.
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Figure 1. A comparison of the calculated and experimental magnetic and nuclear intensities for BaMnF₄, the line indicates \( x = y \) and is a guide for the eye. \[10\]

in figure 1, where a description of the experiment and an example polarization matrix is given in reference [10]. The package also contains routines to draw nuclear and magnetic structures based on the Octave-Forge package ‘vrml’ \[17\]. MuFit is issued under the terms of the GNU General Public License and is freely available. It can be obtained by contacting the author at Bertrand.Roessli@psi.ch.

5. Conclusion
A summary of the essential equations for the description of the intensity of the polarized neutron scattering have been given and a description of how these are used in the MuFit computer package is described. The MuFit package has been successfully used in the analysis of experimental data and has been compared to CCSL. The results from this comparison indicates that the routines give very similar results.

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7. References
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