Developing Wide Angle Spherical Neutron Polarimetry at Oak Ridge National Laboratory

T. Wang¹, N. Silva¹, C. Y. Jiang¹, *, H. K. Agrawal¹, F. Li¹, L. Debeer-Schmitt¹, M. Masaaki¹, J. Ruff¹, R. Pynn², X. Tong¹,³,⁴ and B. Winn¹

1. Neutron Science Directorate, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA
2. Department of Physics, Indiana University, Bloomington, IN 47405, USA
3. Institute of High Energy Physics, Chinese Academy of Sciences (CAS), Beijing 100048, China
4. Dongguan Neutron Science Center, Dongguan 523803, China

* corresponding author email: jiangc@ornl.gov

Abstract. The Spherical Neutron Polarimetry (SNP) Technique probes complex magnetic structures which are inaccessible by other methods. This technique is achieved by combining a zero-field sample chamber with full control of incoming and outgoing neutron polarization. While wide-angle SNP capability has been fully implemented in Europe, it is still in development in the US. At Oak Ridge National Laboratory, we are developing a wide-angle SNP capability suited for multiple neutron beamlines. The design is based on the basic concept of SNP while utilizing high-Tc, superconducting films and Mu-metal. In this article we report our recent development of the SNP capability and its future applications.

Introduction

Polarized neutrons interact with matter through both nuclear and magnetic scattering. The explicit formula, known as the Blume equation, was best elaborated by Blume¹ in 1963. In polarized neutron experiments, the Blume equation can be further demonstrated in matrix form³ as:

\[
\begin{pmatrix}
P'_{x} \\
P'_{y} \\
P'_{z}
\end{pmatrix} =
\begin{pmatrix}
N'N - \vec{M}_1 \cdot \vec{M}_2 - \frac{1}{2} \sigma_{inc} \\
-2\text{Im}[N\vec{M}_2^x] \\
-2\text{Im}[N\vec{M}_2^y]
\end{pmatrix}
\begin{pmatrix}
2\text{Im}[N\vec{M}_1^x] \\
N'N - \vec{M}_1 \cdot \vec{M}_2 - \frac{1}{2} \sigma_{inc} + 2\text{Re}[\vec{M}_1^x\vec{M}_2^y] \\
2\text{Re}[\vec{M}_1^x\vec{M}_2^y] \\
2\text{Re}[\vec{M}_1^y\vec{M}_2^y]
\end{pmatrix}
\begin{pmatrix}
P_x \\
P_y \\
P_z
\end{pmatrix}
\]

(1)

where \(N\) is the coherent nuclear structure factor, \(M\) is the magnetic structure factor, and \(\sigma_{inc}\) is the incoherent scattering cross section. The “⊥” sign indicates that only the component of \(M\) perpendicular to the scattering vector \(Q\) contributes to the scattering process. For this article, we select the coordinate system such that the x axis aligns with the direction of \(Q\), the z axis is perpendicular to the scattering plane, and the y axis completes the Cartesian coordinates.

Eqn. (1) describes the final neutron polarization \(P'\), which can be expressed as the initial polarization \(P\) multiplied by a rotation matrix, plus an additional polarization term produced by nuclear-magnetic interaction or chiral structures. For conventional polarized neutron scattering experiments³, such as the longitudinal or the XYZ method, neutron polarization analyses are limited to the diagonal terms of the polarization rotation matrix so that only the contribution from nuclear, magnetic and incoherent scattering can be determined. To determine more complex magnetic structures, including the nuclear-magnetic interference and the chiral contribution, it is necessary to perform off-diagonal measurements so that each component of the rotation matrix and the additional polarization term can be calculated.
Access to off-diagonal terms is usually not feasible because of the magnetic guide fields around the sample. The typical setup of a polarized neutron scattering experiment is shown in Fig. 1(a), where the incoming and outgoing neutron polarization are defined by the electromagnets around the sample. The sample magnetic field adiabatically connects to the guide fields on each side, which ensures proper neutron polarization transfer but limits the incoming and outgoing neutron polarization to be parallel/antiparallel to each other. To remove this restriction, it is essential to place the sample in a zero magnetic field environment to fully decouple the incoming and outgoing neutron polarization, shown in fig. 1(b), allowing off-diagonal measurements to be performed.

Figure 1. Neutron polarization transfer configuration. (a) Neutron polarization and guide field configuration for a conventional polarized neutron scattering experiment. (b) Neutron polarization and guide field configuration for Spherical Neutron Polarimetry.

For SNP applications, precise neutron polarization control is critical for aligning incoming and outgoing neutron polarization along any direction\(^4\). This control is achieved through a combination of adiabatic and strictly non-adiabatic neutron polarization transitions. For example, the CryoPAD\(^2,4,5\) first developed at ILL uses Niobium superconducting shields to implement a combination of non-adiabatic transition between guided adiabatic polarization transfer and controlled polarization precession. The MuPAD\(^6,7\) developed at FRM-II, on the other hand, uses mu-metal shielding to provide double neutron polarization precession. Both applications utilize precession of neutron polarization which limits the application to monochromatic neutron sources because neutron polarization precession is energy dependent.

As reported in this article, we aim to develop a fully functional SNP capability for wide-angle applications based on an existing small angle SNP system (CryoCUP)\(^8\) to serve the Polarized Triple-Axis Spectrometer (PTAX) as well as the General Purpose Small Angle Neutron Scattering (GP-SANS) instrument at the High Flux Isotope Reactor (HFIR). Application of the technique to the time-of-flight instrument Hybrid Spectrometer (HYSPEC) at the Spallation Neutron Source (SNS) is in the works as well.

Instrument design and simulation
The general design of the SNP device for instruments at HFIR consists of three major components: a zero-field sample chamber, two neutron polarization control arms, and a separate sample environment. A model of these components is shown in Fig. 2(a) with pictures of the assembled zero-field sample chamber and the precession region in Fig. 2 (b) – (c). The device is approximately 1 m long along the neutron path and 1.2 m high with each precession arm extending 30 cm from the center sample position. The extended precession arm structure provides a 4 cm × 4 cm beam size and allows the device to fit on different scattering beamlines at ORNL.
Figure 2. SNP instrument at ORNL. (a) General design of instrument without support structures. (b) Photo of the mu-metal zero-field sample chamber. (c) Photo of precession region within the support structure, which includes the copper heat transfer structure and superconducting coils.

The zero-field sample chamber in this design is achieved by two concentric layers of mu-metal surrounding an Orange Cryostat tail. The inner and outer mu-metal cylinder are each soldered to a long mu-metal tunnel along the incoming and outgoing neutron directions, extending to the incoming and outgoing precession arms. Both layers are also built with sliding slots to allow up to 85° rotation between the incoming and outgoing neutrons for wide angle coverage. While the incoming tunnel is fixed in place during an experiment, the rotation of the outgoing tunnel is driven by the downstream scattering arm on PTAX to cover -5° to 80° scattering angles. For experiments on GP-SANS, rotation of the outgoing tunnel is usually not necessary.

Simulations of the magnetic field within the zero-field chamber are shown in Fig. 3(a) and 3(b). In both simulations we include the mu-metal and YBCO films from the precession region as their presence affects the field distribution. The simulation results show that with the current design less than 0.32 Gauss·cm field integral is generated for each neutron polarization control arm, which corresponds to a neutron polarization precession of 0.75° per angstrom of neutron wavelength. For our current application, the worst scenario occurs at 6 Å, in which case the stray field integral corresponds to a polarization deviation up to 5.9% from the ideal situation. A 3D Hall probe was used to measure the actual field inside the zero-field chamber which shows that the field is below the detection range of the Hall probe, which is 50 mG.
Figure 3. MagNet© simulation result of the zero-field sample region, with the white dashed arrow marking the direction of the neutron. (a) Entire zero field region contour plot when a 7 G environmental external magnetic field (vertical) is applied. (b) Magnetic field flux from the solenoid (3 A) and nutator (4 A) at the beginning of the zero-field chamber. The color indicates the field magnitude and the black arrows mark the direction of the flux.

The neutron polarization control in this design adopts a two-step approach for both the incoming and outgoing sides, with the order reversed for the outgoing side. For the incoming neutron polarization, demonstrated in Fig. 4(a), it is first guided by a solenoid field $B_1$ (20 G) along the $x'$ direction and then adiabatically transferred into the $y'$-$z'$ plane defined by a Meissner shield. The Meissner effect forces the magnetic field $B_2$ (25 G) to be parallel to its surface while the direction within the plane, defined as the polar angle $\theta$ in the Bloch sphere in Fig. 4(b), is controlled by the nutator magnetic field, which can be rotated around the $x'$ axis. The Meissner shield also creates a sharp magnetic field shift on each side, which allows a vertical field $B_3$ to be applied between two Meissner shields. The vertical field $B_3$ is generated by a pair of coils between the two Meissner shields, shown in Fig. 2(c), with its magnitude (-30 G to 30 G) controlled by the current applied. The combination of the controlled magnetic field $B_3$ and the fixed distance between the two Meissner shields creates a controlled neutron polarization precession that defines the azimuthal angle $\phi$. Thus, in the coordinate system defined by $x'$-$y'$-$z'$, through control first of the angle $\theta$ and then $\phi$, the incoming neutron polarization is well defined before entering the zero-field sample chamber. For outgoing neutrons, the order of the neutron precession and adiabatic transition is reversed.
Figure 4. Polarization control strategy for the SNP setup. (a) Magnetic field setup for the incoming neutron. For outgoing neutrons, the direction of the neutron beam is reversed. (b) Incoming neutron polarization movement on a Bloch sphere when a neutron travels through the magnetic field setup in (a).

The Meissner shield in this development is created by high-T$_c$ superconducting thin films. Two types of films are tested and implemented in this design. For current applications at PTAX and GP-SANS, where each measurement only needs to cover a small angle, the flat YBCO thin film (200 nm) deposited on a single crystal sapphire (7 cm×10 cm), shown in Fig. 5(a), is used. The performance of this type of YBCO film has been measured in previous work\textsuperscript{9,10,11} which demonstrated that they provide a reliable Meissner effect below its critical temperature of 90 K with a 98.4% neutron transmission. The other type of film is the thin YBCO film deposited on an Yttria-stabilized Zirconia (YSZ) substrate which has a bending radius of 75 mm and a critical temperature around 90 K. The bendable nature of the YSZ film provides the possibility to cover a wide angular range simultaneously while still maintaining a relatively high functioning temperature compared to the niobium used in CryoPAD. The feasibility of the YSZ film has been tested by Ceraco\textsuperscript{©} with large quality-stable production pending. In a recent test experiment at HFIR, it was demonstrated that a larger continuous Meissner shield can be formed by offset-stacking multiple superconducting thin films\textsuperscript{12}. As of this publication, both bendable and offset stacks method are in consideration for time-of-flight instruments. To keep the superconducting film under its critical temperature, a Sumitomo CH-110 single stage closed-cycle refrigerator is used, which provides a cooling power of 200 W at 77 K. To further reduce heat generation, the coils for the precession field $B_3$ are made by YBCO superconducting tapes (Superpower\textsuperscript{©}).

Figure 5. Pictures of superconducting thin films. (a) YBCO films (200 nm) deposited on single crystal sapphire substrate and covered with a gold protecting layer. (b) YBCO film deposited on bendable YSZ substrate.

The sample temperature control is achieved by a customized orange cryostat compatible with the zero-field mu-metal shielding structure. The customization is applied to the tail of the orange cryostat where it is narrowed to 80 mm outer diameter with a depth of 232.5 mm to fit in the tall cylinder-shaped mu-metal zero-field chamber. Despite the narrow outer diameter, the orange cryostat still provides a 50 mm inner diameter sample space and is equipped with a non-magnetic goniometer (AttoCube© ANGt10) to allow in-situ sample alignment. The temperature range provided by the orange cryostat is 1.5 K – 350 K, operating separately from the neutron polarization control units.

Conclusion and future experiment

Currently the SNP device is being assembled which will be followed by several offline tests such as the cooling and the vacuum test. The first online test and calibration is scheduled at the HFIR HB-2D beamline to tune the whole setup. As a demonstration experiment, the new capability will be used to examine the multiferroic sample MnWO$_4$ at the HFIR PTAX beamline. For HYSPEC, a different design needs to be
implemented because of its time-of-flight nature. Currently, a couple of design concepts are being considered and will be finalized in early 2019.

Acknowledgement
This work was supported by the U.S. Department of Energy (DOE), Office of Science (OS), Basic Energy Sciences (BES), Materials Sciences and Engineering Division (sample design, fabrication, and physical property characterization) and by the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory (ORNL), managed by UT-Battelle, LLC, for the U. S. DOE (PNR). The research at ORNL’s High Flux Isotope Reactor was sponsored by the Scientific User Facilities Division, BES, U.S. DOE. The superconductor used in this work is a collaboration with Dr. Robert Semerad at Ceraco®.

References
1. M. Blume, Phys Rev 130 (5), 1670 (1963).
2. E. Lelievre-Berna, P. J. Brown, F. Tasset, K. Kakurai, M. Takeda, and L. P. Regnault, Physica B 397 (1-2), 120 (2007).
3. O. Scharpf and H. Capellmann, Phys Status Solidi A 135 (2), 359 (1993).
4. F. Tasset, Physica B 156, 627 (1989).
5. M. Takeda, M. Nakamura, K. Kakurai, E. Lelievre-berna, F. Tasset, and L. P. Regnault, Physica B 356 (1-4), 136 (2005).
6. V. Hutanu, W. Luberstetter, E. Bourgeat-Lami, M. Meven, A. Sazonov, A. Steffen, G. Heger, G. Roth, and E. Lelievre-Berna, Rev Sci Instrum 87 (10) (2016).
7. T. Wang, S. R. Parnell, W. A. Hamilton, F. Li, A. L. Washington, D. V. Baxter, and R. Pynn, Rev Sci Instrum 87 (3) (2016).
8. T. Wang, F. Li, S. R. Parnell, W. A. Hamilton, H. Kaiser, A. L. Washington, D. V. Baxter, and R. Pynn, J Phys Conf Ser 528 (2014).
9. S. R. Parnell, H. Kaiser, A. L. Washington, F. Li, T. Wang, D. V. Baxter, and R. Pynn, Physcs Proc 42, 125 (2013).
10. F. Li, S. R. Parnell, T. Wang, D. V. Baxter, and R. Pynn, International Conference on Polarised Neutrons for Condensed Matter Investigations (Pncmi 2014) 711 (2016).
11. T. Wang, C. Y. Jiang, H. Z. Bilheux, J. C. Bilheux, L. Crow, L. Robertson, R. J. Taylor, R. Pynn, S. Parnell, W. A. Hamilton, and X. Tong, Appl Phys Lett (in preperation).