Journal of Neutron Research, Vol. 13, No. 4, December 2005, 193–223

Polarized neutrons for pulsed neutron sources

IAN. S. ANDERSON†, JEREMY COOK‡, GAIN FELCHER§, TOM GENTILE‡,
GEOFFREY GREENE¶, FRANK KLOSE†, TOM KOETZLE§, EDDY LELIEVRE-BERNA††, ANDRE PARIZZI†, ROGER PYNN*‡‡ and JINKUI ZHAO†

†Sapllation Neutron Source, Oak Ridge National Laboratory, Oak Ridge, TN 37831,
‡National Institute of Standards and Technology, Gaithersburg MD 20899,
§Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439,
¶Department of Physics, University of Tennessee, Knoxville TN 37996,
††Institut Laue-Langevin, BP 156, Grenoble Cedex, 38042, France,
‡‡Los Alamos National Laboratory, Los Alamos, New Mexico 87545.

(Revised 12 April 2005)

Polarized neutrons provide a powerful tool for research at today's continuous neutron sources, permitting unique information to be obtained from neutron-scattering experiments and in the area of fundamental neutron physics. The information obtained is indispensable to important areas of current research such as magnetism, spin fluctuations in correlated-electron materials, nanoscience, astrophysics, and cosmology.

Unfortunately, the polarized neutron capabilities that already exist at reactor neutron sources, mainly in Europe, cannot be transferred directly to pulsed spallation sources because many of the devices used at reactors to manipulate neutron spins operate only at a single neutron wavelength or with a neutron beam of limited divergence. Both of these restrictions must be overcome for devices to be useful at pulsed spallation sources. This report identifies the significant R&D efforts that will be required to achieve these goals. These efforts must be undertaken now, if they are to be useful early in the lifetime of the spallation neutron source (SNS) or its Japanese counterpart, Japanese proton accelerator research complex (J-PARC).

A workshop on the use of polarized neutrons at pulsed neutron sources was held on February 10–13, 2003, in Gaithersburg, Maryland, USA. Sixty-one scientists representing work in Europe, Japan and the US attended the workshop whose purpose was to discuss research opportunities with polarized neutrons at pulsed spallation sources and to develop a roadmap for the technology R&D required to facilitate this type of work.

Well-funded efforts to develop polarization capabilities are under way in both Japan and Europe. The workshop concluded that it makes sense for the US to collaborate with these activities, as well as to develop its own R&D program, as it is beginning to do. There is ample opportunity within the R&D portfolio outlined in this report for research activities both at universities and at national neutron centers. Synergy between these efforts will contribute to other goals such as the development of a robust programme of instrument innovation that will be required at new pulsed sources to maintain their vitality in future decades.

Keywords: Polarized neutions; Pulsed neution source; Neution scattering; Neutron spin echo

---

*E-mail: rpynn@indiana.edu

Journal of Neutron Research
ISSN 1023-8166 print/ISSN 1477-2655 online
http://www.tandf.co.uk/journals
DOI: 10.1080/10238160500135673

1. The scientific case for polarized neutrons in neutron-scattering experiments

The use of spin-polarized neutrons in neutron-scattering experiments (at either continuous or pulsed neutron sources) provides the capability to study scientific phenomena and/or achieve levels of instrument performance not otherwise accessible.

The first of these advantages is based on the fact that both the nuclear and magnetic potentials experienced by neutrons interacting with condensed matter depend strongly on the spin polarization of the neutron. This polarization dependence can be exploited to extract quantitative information about spatial and temporal variations of atomic and magnetic densities in condensed matter that cannot be obtained with unpolarized neutrons. Polarized neutrons thus make it possible to study qualitatively different scientific phenomena from those measurable with unpolarized neutrons.

The second part of the case relies on the fact that manipulations of neutron spins before or after scattering from a sample—for example, Larmor precession of neutron spins in an applied magnetic field—can be used to "label" the speed or trajectory of each neutron, thereby eliminating the need to define these quantities using conventional collimators or monochromators that cause significant loss of signal intensity. The best-known example of this situation is neutron spin echo (NSE) spectroscopy, a technique that provides the best energy resolution obtainable for neutron inelastic scattering experiments.

It is important to note that the scientific opportunities described above can, at least in principle, be realized at either pulsed or continuous neutron sources, provided they are sufficiently powerful. All that is required are technologies for polarizing neutrons and for manipulating them. A principal goal of the workshop on which this article is based was to discuss polarized-neutron technologies appropriate for pulsed spallation neutron sources. These technologies are not yet sufficiently developed to allow pursuit of the scientific agenda described below.

In this section, we describe each part of the scientific case for polarized neutrons separately, after first digressing to place them in an appropriate historical context.

1.1 Historical perspective

The first work with polarized neutrons began four years after the 1932 discovery of the neutron by Chadwick [1], with the suggestion by Bloch [2] that polarized neutron beams could be produced by transmission through magnetized iron. In 1939, Halpern and Johnson [3] published an article that has served as the basis for all subsequent theoretical and experimental work.

The construction of nuclear reactors provided more intense neutron sources, and postwar experiments carried out at Argonne and Oak Ridge made possible the production and use of polarized monochromatic beams of neutrons. By the early 1950s, there were three proven methods for producing polarized neutron beams: (1) transmission through magnetized iron, (2) total reflection from magnetized mirrors, and (3) Bragg scattering from ferromagnetic single crystals. The first two-axis polarized beam diffractometers were built at Brookhaven and the Massachusetts Institute of Technology. $Co_{0.92}Fe_{0.08}$ single crystals and resonant flippers were rather quickly adopted at other neutron-scattering centres to measure magnetization distributions.

In the 1960s, the theory of polarized neutrons scattered by various materials was developed by Izyumov and Maleyev [4], Blume [5,6], Izyumov [7], and Schermer and Blume [8]. The theoretical results gave complicated expressions for the total cross section and the final polarization vector in terms of the initial polarization and the nuclear and magnetic scattering amplitudes.

In 1969, Moon, Riste, and Koehler [9] at Oak Ridge studied the various effects that are present in the scattering of polarized neutrons and introduced the four spin-flip and non-spin-flip cross sections that can be deduced by keeping only the neutron spin component parallel to the incident polarization. The fact that the count rates observed by these authors were so low, even at an 85 MW reactor and for "fruit fly" samples, discouraged the use of polarized neutrons for all but the simplest application (i.e., measurement of atomic form factors) for more than a decade. Even though Moon, Riste, and Koehler recognized that their application of a constant magnetic guide field restricted the number of independent magnetic cross sections that could be measured, this point was missed by many and slowed the development of generalized polarization analysis (GPA).

In 1971, Mezei [10] demonstrated the spin echo method that allowed very small changes of neutron velocity to be observed independently of the velocity spread in the neutron beam. Mezei and scientists in Russia also invented supermirrors [11] that increased the angular divergence and wavelength range of neutron beams that could be polarized by mirror reflection. With the successful development of supermirrors at the Institut Laue-Langevin (ILL), Schaerpf [12] reconstructed the diffuse-scattering instrument D7 to accommodate polarization analysis. A huge intensity gain was obtained by using 32 detectors equipped with 3200 supermirror analyzers (more such analyzers have been added over the years, and D7 now has more than 6000 supermirrors). Three mutually perpendicular Helmholtz pairs at the sample position allowed the polarization of the incident and scattered neutron beams to be aligned with any of the three directions x, y, or z (three-directional polarization analysis). From the six measured cross sections, one can separate the nuclear, nuclear spin incoherent, and magnetic contributions. This is a generalization of the technique studied previously by Moon, Riste, and Koehler (single-directional polarization analysis).

The 1964 discovery by Brown and Forsyth [13] that the (111) reflection of the Heusler alloy Cu_2MnAl is perfectly polarized eventually led in the early 1980s to the development of polarizing monochromators that could focus polarized neutrons on a sample and provide enough intensity for inelastic neutron-scattering measurements to be made with polarization analysis. The key in this case was careful identification and extraction of single-crystal grains from a Heusler alloy ingot [14] and their deployment in a vertically focusing monochromator. Neutron spin flippers were also improved during this period. The radio-frequency (rf) coils proposed by Shull and used by Moon, Riste, and Koehler were replaced by DC coils developed independently by Mezei [15] and Rekveldt [16]. Tasset proposed and built a third type of flipper called a Cryoflipper, in which two opposite magnetic fields are separated by a thin Meissner sheet.

In the 1980s, several scientists in Europe and Japan developed instrumentation based on the pioneering experiment of Alperin [17], who had demonstrated that GPA could be realized by connecting two different guide-field directions onto a zero-field sample chamber. The most ambitious of these efforts was undertaken by Tasset [18], who built an apparatus at ILL to determine the direction of the scattered polarization vector for any given incident polarization and any scattering angle. Using his expertise with superconducting screens developed from building the Cryoflipper, he constructed a compact cryogenic polarization analysis device (Cryopad) that takes advantage of the Meissner shields to define the magnetic field and zero-field regions crossed by the incident and scattered neutron beams. Thanks to this device, all the components of the complicated expression of the final polarization vector can be measured, providing unique information on magnetic structures and nuclear/magnetic interferences occurring in the neutron-scattering process.

Because both mirror and single-crystal neutron polarizers have limitations, alternative methods for polarizing neutron beams have been discussed for many years. A project to build ^{3}He spin polarizers was first discussed in 1981 at ILL, and some years later, thanks to progress made in gas polarization for other purposes, a group from ILL and Harvard University [19] carried out an experiment of optical pumping of rubidium vapor and Rb-^{3}He collisions at room temperature. The principle of this polarizing filter is based on the enormous difference in the absorption cross sections for neutrons with spin parallel and antiparallel to the spin of a ^{3}He nucleus first measured in 1966 by Passell and Schermer [20]. In 1996 at ILL, Humblot *et al.* [21] successfully tested an apparatus constructed at Mainz University, which includes optical pumping and compression of the ^{3}He. In this case, the gas was polarized by optical pumping of ^{3}He atoms excited by an electric discharge at ~ 1 mbar and collisions of excited ^{3}He—^{3}He nuclei. Since then, several experiments have been successfully carried out at ILL, and a second-generation ^{3}He filling station has already given impressive results.

From the foregoing discussion, it is obvious that almost all of the pioneering work with polarized neutrons has been carried out at steady-state neutron sources. Although techniques such as polarized neutron reflectometry first developed at pulsed sources, most of the methods that use polarized neutrons have not yet made the transition from continuous to pulsed sources.

1.2 Separating coherent and spin-incoherent nuclear scattering using polarized neutrons

The interaction potential of a neutron and an atomic nucleus depends on the spin state of the compound nucleus formed during the interaction. As a result, the scattered neutron intensity separates into two components—called coherent and (nuclear spin) incoherent scattering—which affect neutron polarization differently. *Coherent* scattering results in no change of the neutron polarization during scattering, while the *incoherent* process results in two-thirds of the scattered neutrons having their spins flipped during the scattering process (i.e. the polarization of the scattered neutron beam is -0.33). This difference in behavior immediately provides a way in which coherent and incoherent scattering can be separately identified, at least in nonmagnetic samples. Since coherent nuclear scattering of neutrons measures correlations between the (time-dependent) positions of *distinct* nuclei while incoherent scattering reflects *single-nuclei* correlations, such a separation can be important in identifying the physical processes occurring within the scattering sample. For example, *incoherent* quasielastic neutron scattering provides information about atomic jumps and diffusion, whereas *coherent* inelastic neutron scattering results from correlated movements of separate atomic nuclei, such as those caused by phonon propagation.

1.3 Studying magnetism with polarized neutrons

Neutrons have a magnetic moment (which is aligned antiparallel with their spin) and are thus sensitive to space- and time-dependent magnetization fluctuations in solid samples. Because of the dipolar nature of this magnetic interaction, only the component, $\vec{M}_\perp$, of sample magnetization that is perpendicular to the neutron wavevector transfer $\vec{Q}$ is effective in scattering neutrons. This dependence makes it possible in some cases (notably in isotropic ferromagnets) to separately measure the magnetic neutron scattering by applying a saturating magnetic field to the sample and taking the difference between scattering obtained with the field perpendicular and parallel to $\vec{Q}$. The use of polarized neutrons, however, makes the separation of magnetic and nuclear scattering much easier because scattering by fluctuations in $\vec{M}_\perp$ that are parallel to the quantization direction of the neutron spins do not affect the spin direction of the neutron (giving rise to so-called non-spin-flip scattering). On the other hand, fluctuations in $\vec{M}_\perp$ that are perpendicular to the neutron quantization direction cause a change in the direction of the neutron's spin (causing so-called spin-flip scattering in simple 1-d polarization analysis experiments). This dependence of magnetic scattering on the initial and final spin states of the neutron has been used in many experiments and probably provides, on its own, a justification for the production and use of polarized neutrons in neutron-scattering experiments. It has been used to deduce the spatial distributions of magnetization as well as the direction of the magnetization vector on an atomic scale in a wide variety of samples, some of which are described below. It has also played a crucial role in supporting the development of new magnetic materials.

1.4 Polarized neutron scattering by molecular and organic magnets

Research in magnetic materials has exploded in recent years because of the development of new molecular and organic magnets, that is, solids that are built up from structurally well-defined clusters containing magnetic ions in a complex environment [22]. Since the discovery of the first ferromagnetic molecular compound (decamethylferrocenium tetracyanoethylenide, $T_C = 4.8\,\mathrm{K}$) in 1986, enormous progress has been made in this area. These molecular magnets are typically polynuclear transition metal complexes and they can be termed “single molecule magnets.” The unpaired electron responsible for the magnetism sits in a molecular orbital built up from the orbitals of the atoms constituting the molecule. The magnetization tends to be smeared out across the molecule, though perhaps concentrated on certain atoms. Measuring the magnetization distribution across a molecule reveals precious information on the nature of the molecular orbitals responsible for the magnetism and the interactions with neighbouring molecules in the solid, as well as the chemical bonding and how the electron spin is spread out and oriented. This allows for testing the underlying theories of molecular bonding and magnetism and for creating new magnetic materials with predicted properties. Molecular magnets can also be viewed as single-domain magnets with a domain size in the nano limit. Thus, they can be used for studies of magnetic phenomena on the nano scale.

A typical example of a molecular magnet is the room-temperature magnet combining a hexacyanometalate $[\mathrm{M(CN)_6}]^{q-}$ with a Lewis acid L^{p+} [23] (see figure 1). If L and M are transition metal ions, the orbital interactions in the resulting compound can be described by

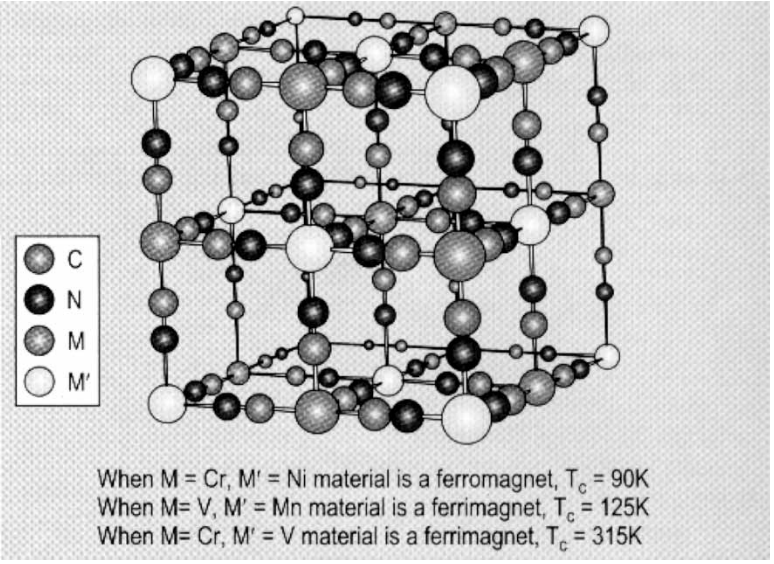

Figure 1. Nuclear structure of the hexacyanometalate $[\mathrm{M(CN)_6}]^{q-}$ with a Lewis acid L^{p+}.

well-understood principles, and it is therefore possible to tune the compound's magnetic properties.

These compounds are of great scientific interest, but a major driving force in this work is the urgent need to find new applications that will exploit their specific properties such as lightness, transparency, solubility, optical properties, and biocompatibility. Molecular magnets with total spin number $S = 10$ also display numerous excited spin states and open the way to a novel class of information storage systems.

Studies of the magnetic properties of such systems aim at understanding both the materials properties (e.g. magnetic coupling mechanisms) and the fundamental chemistry, such as reactivity in coordination complexes and its dependence on electronic structure. An area of research where neutron diffraction has not yet shown its full potential, because of the flux limitations of the present sources, is to combine charge density analyses with magnetic studies in order to obtain an explanation for various magnetic phenomena in terms of electronic structure. An example where this would be useful is the large molecular magnet or "chromium wheel" system, $Cr_8F_8(C_5H_9O_2)_{16}$, containing 272 unique atoms [24]. For this structure, the charge density has been determined from synchrotron X-ray data, and a detailed topological analysis of the electron density has been carried out. Figure 2 shows the experimentally determined electrostatic potential, which is used for predicting the inclusion properties of the molecule. A combination of magnetic neutron-diffraction data and synchrotron X-ray data could provide electronic information on complex molecular systems such as this, which is very difficult to obtain by other methods (e.g. theoretical calculations).

In the area of molecular magnetism, the number of studies of organic radicals has also increased dramatically. Even though radicals are chemically reactive, there are many examples of materials with radicals trapped in the solid phase, such as nitroxides [25]. These

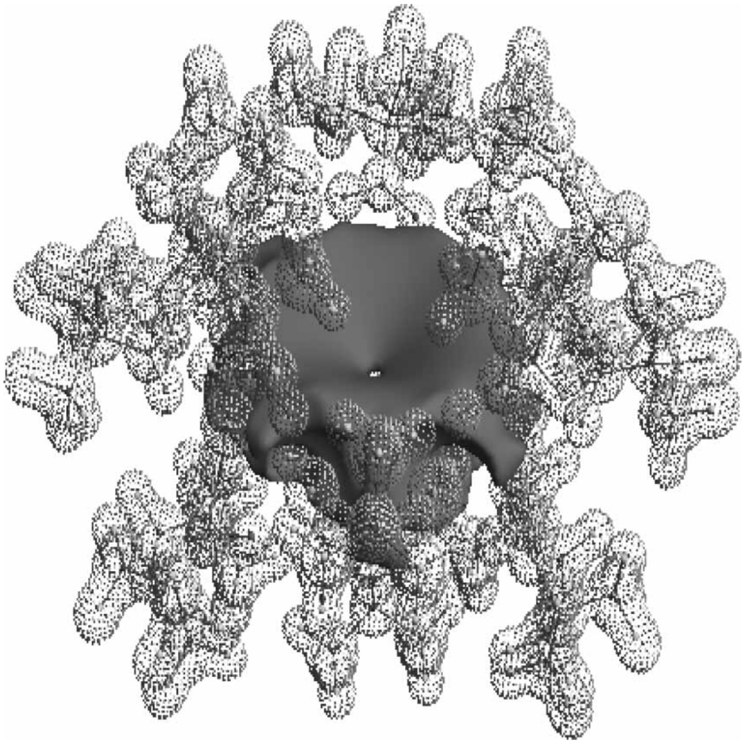

Figure 2. The experimentally determined electrostatic potential of the "chromium wheel" [Cr8F8(C5H9O2)16]. Surface at $-0.54\,e\,\mathring{A} - 1$ in red and $+0.30\,e\,\mathring{A} - 1$ in yellow.

materials exhibit many different magnetic phenomena, which to a large extent are determined by the crystal packing and the detailed nature of the intermolecular interactions. Thus, the field of organic radicals will be particularly well suited for research with next-generation neutron sources such as SNS.

As the previous discussion shows, neutron diffraction remains the technique of choice for studying magnetism. It is the classical polarized neutron-diffraction technique that permits investigation of the distribution of the magnetization, which contains essential information on the electronic structure of materials: the nature of the magnetic orbitals, the interactions with neighboring molecules in the solid, and effects such as chemical bonding, spin delocalization, or spin polarization. Up to now, this technique has only been used at continuous neutron sources such as the ILL (using the diffractometer D3) because there has been no device able to simultaneously polarize a beam efficiently at short wavelengths while maintaining the bandwidth. Today, thanks to the progress made in the development of polarizing guides and ^{3}He neutron spin filters (see later sections on techniques), one can envisage applying classical polarized beam diffraction at a pulsed neutron source.

1.5 Magnetic moment configurations determined by polarized neutron scattering

The determination of magnetic structures plays a major role in the understanding of phenomena such as low-dimensional systems, phase transitions, and geometric frustrations. This determination is far from trivial, however. Even a “simple” material like elemental neodymium is fascinating. During the past 40 years or so, enormous effort has gone into understanding the electronic and magnetic properties of neodymium, yet the great majority of the antiferromagnetic structures that are stabilized below 20 K under zero or applied field remain unsolved.

In the case of powder diffraction, many Bragg peaks are superimposed in the measured patterns, and Rietveld refinements are rarely able to determine a magnetic configuration unambiguously, except in special cases where there are many extinction rules implying severe constraints [26]. In a single-crystal experiment, the time-of-flight technique provides large Laue maps of reciprocal space, and multidomain/single-*k* structures can be distinguished from multi-*k* ones. As an example of such a study, figure 3 shows the

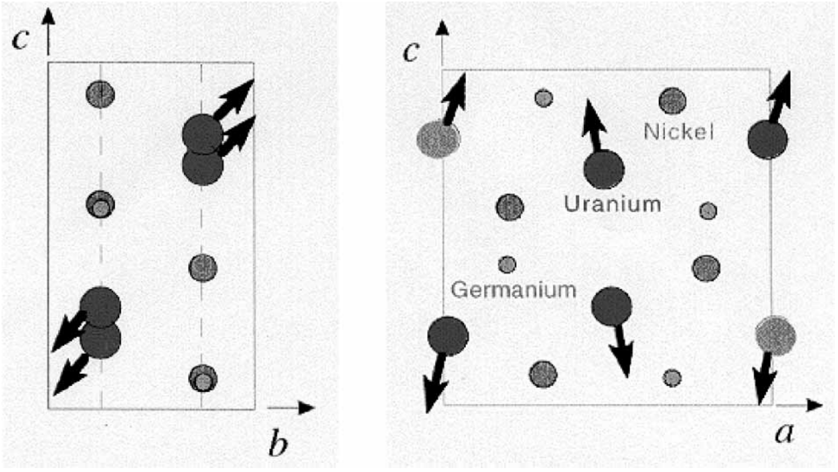

Figure 3. Moment configuration of one domain in UNiGe shown as projections on the orthorhombic b-c and a-c planes, respectively [27]. The moment directions of the second domain are given by the images at the $y = 1/4$; and 3/4; mirrors (dashed lines).

proposed moment configuration in UNiGe. For this orthorhombic compound, time-of-flight, single-crystal neutron diffraction revealed a complex configuration of uranium 5f moments, with a single-*k* propagation vector, and the occurrence of two magnetic domains below $T_N = 42\,\mathrm{K}$ [27]. In general, single-crystal neutron diffraction makes it possible to separate the nuclear and magnetic contributions to the Bragg peaks [28] and to distinguish coexisting magnetic phases in a single material [29]. With the presence or absence of key reflections, magnetic configurations of collinear structures are generally determined unambiguously, but for complex antiferromagnets, polarized neutron techniques generally must be employed.

For magnetic structure investigations requiring an applied magnetic field, the transverse components of the neutron polarization are inaccessible, but selection rules can be applied. When the orientation of the single crystal is suitably chosen, relative to the field (or polarization vector) and the scattering vector, one measures spin-flip and non-spin-flip cross sections that reveal some of the information inaccessible from integrated intensity measurements alone. For example, one can determine the component of moments that are transverse to the applied field. This technique was used to confirm the noncollinear magnetic structure of UNiGe shown in figure 3 [28].

1.6 The interplay between charge, magnetic, and lattice fluctuations

Since the advent of quantum mechanics, the field of magnetism has attained a special status as an arena in which to develop and test new theories and ideas. The last decade has seen an intensified interest in low-dimensional systems, where quantum fluctuations are germane. The cooperative phenomena of macroscopic quantum ground states and quantum phase transitions, where order is destroyed by quantum fluctuations, have been studied both theoretically and experimentally. Materials where localized magnetic moments are arranged and coupled in specific ways have provided experimental insight into the behaviour of many-body quantum systems. In particular, a large group of such model systems have been found in materials with spins from unpaired electrons on metal ions coupled by super-exchange interactions through connecting oxygen ions (cupreates, vanadates, nickelates, etc.).

It has become clear that several novel phenomena in condensed matter physics, such as high-temperature superconductivity or giant magneto-resistance, require an extension of the localized electronic-moment picture. Common features of these novel phenomena are that a major role is played by the quantum fluctuations, but the understanding of quantum effects in purely magnetic systems must be extended to include orbital effects, charge fluctuations, and lattice distortions, which can be either long-range ordered, correlated on short-length scales, or just dynamic fluctuations. The understanding of these materials involves a competition between charge, spin, and orbital and lattice degrees of freedom both on a static and on a dynamic level. Such a competition should generate “hybrid” correlation functions coupling the various degrees of freedom (charge-lattice, charge-magnetic, spin-lattice, spin-orbit, etc.), which can be accurately measured using powerful inelastic or quasi-elastic polarized neutron-scattering techniques such as GPA. Indeed, by allowing the measurement of the transverse components of polarization, this method gives unique access to the so-called inelastic magnetic-nuclear interference terms. These terms could provide interesting information about the hybrid pair correlation functions, which play a crucial role for the understanding of strongly correlated electron systems [30].

1.7 Nanomagnetism

The world is entering an era in which manipulation of charge and spin offers the possibility to replace present-day semiconductor electronics, just as vacuum tube electronics were supplanted in the past. The term coined to embrace the new wave is "spintronics." Ultimately, the goal will be to transcend binary logic and move toward quantum computing strategies that can be implemented via electronic or nuclear spin manipulation using quantum-entanglement. Spintronics can converge with the burgeoning field of molecular electronics toward this end. However, there are many challenges ahead. For example, the opportunity to fabricate new systems on length scales that compete with those relevant to magnetism will challenge the fundamental knowledge in magnetism and naïve wisdom that magnetic properties at the nanoscale can be understood in terms of bulk or atomic magnetic properties. In addition, control of magnetism at the nanoscale offers a pathway to create new devices using systematic principles of nanotechnology.

The role of neutron scattering and other techniques in the study of nanostructured magnetic materials has recently been examined in detail in a review article by Fitzsimmons, *et al.* [31] These authors point out that the key issues in these technologically promising materials is to relate their physical properties (transport, magnetism, mechanical, etc.) to their chemical and physical structure. Success in this endeavour requires detailed quantitative understanding of magnetic structure and properties (which polarized neutrons can often provide), as well as the development of new modeling and simulation capabilities. Progress in applying neutron-scattering methods to samples of ever decreasing size has allowed the technique to be applied to nano-structured materials prepared by thin-film and lithographic techniques. Among the interesting results that have been obtained, Fitzsimmons *et al.* note: distinguishing between magnetic and chemical boundaries; observing the spatial dependence of the magnetization vector in nonuniform materials; unusual coupling mechanisms across nonmagnetic materials; and unexpected magnetic phase diagrams. These authors anticipate that the extension of elastic neutron scattering to nanostructured arrays and three-dimensional magnetic composites will allow future determination of magnetic structure with unprecedented resolution.

Polarized neutron reflectometry (PNR) has been used very successfully to investigate nanostructured systems composed of thin layers where it has elucidated magnetization profiles close to surfaces and interfaces. Surface sensitivity derives from working in grazing incidence geometry near the angle for total external reflection. Polarized neutron reflectometry is highly sensitive, capable of measuring the absolute magnetization of a monolayer of iron ($\sim 10^{-4}$ emu) with 10% precision and has excellent depth resolution—on the order of a tenth of a nanometer even for films as thick as several hundred nanometers. Polarized neutron reflectometry has enjoyed dramatic growth at both steady-state and pulsed neutron sources during the last decade and has been applied to important problems such as the origin of exchange bias, magnetic reorientation transitions in thin films, enhanced magnetization at surfaces and interfaces, magnetic penetration in superconductors, and the nature and importance of magnetic roughness. Each of these areas, as well as the opportunities for future research involving the use of polarized neutrons, has been examined in detailed by Fitzsimmons *et al.* [31]

1.8 Time-dependent magnetic fluctuations in materials

Neutron polarization analysis for inelastic scattering experiments was first seriously developed at ILL in the early 1980s by adding polarizing devices such as Heusler alloy monochromators and analyzers, as well as supermirror benders, to classical thermal and cold neutron three-axis spectrometers. The new spectrometers—IN12, IN20, and IN14—proved so successful that they have subsequently been duplicated in North America, Japan, and several European countries. Among early successes at ILL were verification of the Haldane conjecture [32], determination of the spectral form for spin diffusion in isotropic ferromagnets [33], identification of new, coupled modes in one-dimensional magnets [34], and studies of nonlinear, solitary-wave dynamics in both ferro and antiferromagnetic low-dimensional systems [35,36]. More recently, inelastic scattering with polarization analysis has been applied successfully to the study of the spin-dynamics of high-temperature superconductors [37] and other correlated-electron systems [38].

1.9 Generalized polarization analysis

Until a few years ago almost all neutron-scattering experiments with polarization analysis used only one component of the beam polarization. A polarized beam of neutrons was prepared in which the spins of all neutrons were aligned along an applied magnetic field. Only those spin components of the scattered neutrons that were either parallel (+ direction) or antiparallel (− direction) to the applied field were measured, allowing a total of four different cross sections (++, − −, + − and − +) to be measured. Even though this spin-projection method is very powerful, and enables all of the measurements described in previous paragraphs, it does not exhaust all of the information about sample magnetism that can be obtained using polarized neutrons. In general, the magnetic response of a solid to an applied field is described by a second-rank susceptibility tensor that describes how the α Cartesian component of the field affects the β component of the magnetization. By the fluctuation-dissipation theorem, the susceptibility tensor is related to scattering cross sections for neutrons in which the spin directions of incident neutrons also lie along different Cartesian axes. Measuring such cross sections is, however, not straightforward and can only be done if the sample has no macroscopic magnetization and can be placed in a region of zero magnetic field. In such circumstances, GPA is possible because the spatial orientation of the spins of both the incident and scattered neutrons can be controlled. GPA (sometimes also known as "spherical polarimetry") is particularly useful for studying magnetization fluctuations in materials in which atomic moments are non-colinear.

With this technique it has been possible to solve a number of magnetic structure problems that had proven to be intractable when employing other techniques [39]. Very recently, the technique has been applied successfully to the determination of antiferromagnetic densities. When the magnetic and nuclear scatterings occur at the same place in reciprocal space, this method enables the precise determination of antiferromagnetic form factors, which is not possible by other means. For example, the investigation of the magnetization distribution of Cr_2O_3 has revealed that the Cr^{3+} magnetic moment is reduced by the zero-point spin deviation and by covalent mixing to $2.48\,\mu_B$ [40]. These results are consistent with the chromium d electrons being in the trigonally symmetric a_1 and e orbitals derived from the cubic orbitals with t_{2g} symmetry. There is a small but significant magnetization that is not accounted for by these orbitals and which is attributed to covalent overlap. Its symmetry is consistent with the magneto-electric susceptibility. Very recently, V. Fedorov and coworkers have also proposed a new method that takes advantage of GPA, based on the passage of cold neutrons through non-centrosymmetric single crystals in Laue diffraction, to search for the neutron electric dipole moment. The sensitivity of the method relies on the high interplanar electric field in non-centrosymmetric crystals (up to 10^9 V/cm) and on the possibility to increase the time the neutron spends in the crystal by using large Bragg angles.

2. Neutron polarization as a tool to enhance neutron-scattering instrumentation

The second part of the scientific case for polarized neutrons in neutron-scattering experiments involves their use in improving the performance of neutron-scattering spectrometers. In general, spectrometers that use polarized neutrons in this way are not designed to exploit the dependence of neutron-scattering cross sections on neutron polarization but rather make use of the neutron magnetic moment as a spectrometer design element. The most widely used instrument design concept using polarized neutrons is the NSE method invented by Mezei in 1972 [10]. In this method, neutron spins undergo Larmor precession in magnetic fields placed before and after a scattering sample. These fields are arranged so that each neutron will experience an equal and opposite number of precessions before and after scattering, whatever the neutron's actual velocity, provided the sample scattering is elastic. At the echo position, all of the precessing neutron spins are in phase, whatever the neutron velocity, and the beam is fully polarized. If the scattering is inelastic, the numbers of neutron spin precessions before and after scattering are not equal, leading to depolarization of the neutron beam at the echo point. The depolarization turns out to be a direct measure of the difference between the incident and scattered neutron velocities and is independent to the lowest order of the actual neutron velocity. The NSE method thus provides a sensitive measure of neutron velocity changes that occur during scattering, to a large extent independently of the degree of beam monochromatization. For example, a neutron beam with a 10% velocity spread can be used with the NSE method to measure inelastic neutron scattering with an energy resolution that is substantially less than 0.1% of the incident neutron energy. Conventional methods of achieving good energy resolution, which involve defining accurately both the incident and scattered neutron energies, lead to decreases in scattered neutron intensity that usually have to be compensated by a corresponding degradation of Q resolution. A striking example of this is backscattering, in which huge banks of analyzer crystals are used. Because it breaks the usual relationship between beam monochromatization and energy resolution, the NSE method allows excellent energy resolution and reasonable measured neutron intensity to be achieved simultaneously. Although NSE has been exploited only at steady-state neutron sources since its invention, recent developments at ILL using chopper modulation of the incident neutron beam on the IN15 spectrometer have demonstrated that NSE can be used in time-of-flight mode, paving the way for the use of the technique at pulsed neutron sources.

The NSE technique has proven remarkably useful in the study of polymers and other complex fluids, as well as in determining the slow dynamics associated with glasses [41]. It has even been applied successfully to determine the lifetime of collective excitations in superfluid helium, where it extended by several orders of magnitude the phonon and roton line widths measured by more traditional neutron techniques.

As long ago as 1979, Mezei [42] and Pynn [43] described ways in which NSE could be used to measure the energy widths of collective excitations such as phonons or to improve the angular resolution in diffraction experiments. Because of the difficulty of designing the magnetic field regions needed to implement these ideas, practical applications had to await the development of the so-called neutron resonant spin echo (NRSE) technique that was first demonstrated in Germany by Golub and his collaborators [44]. Although, like NSE, this technique also makes use of Larmor precession of the neutron's spin to code some (vector) component of the neutron velocity, it does so using small coils that produce RF magnetic fields in well-defined regions of the neutron beam line. In this case, it is the RF frequency and the distance between neighboring RF coils that determines the attainable instrumental energy resolution, rather than the strength and spatial extent of the static magnetic fields applied in the standard NSE method. The NRSE method for measuring shapes of dispersive excitations has now been installed on two three-axis spectrometers at steady-state neutron sources in Germany (at HMI and in Munich) and is beginning to produce interesting scientific results.

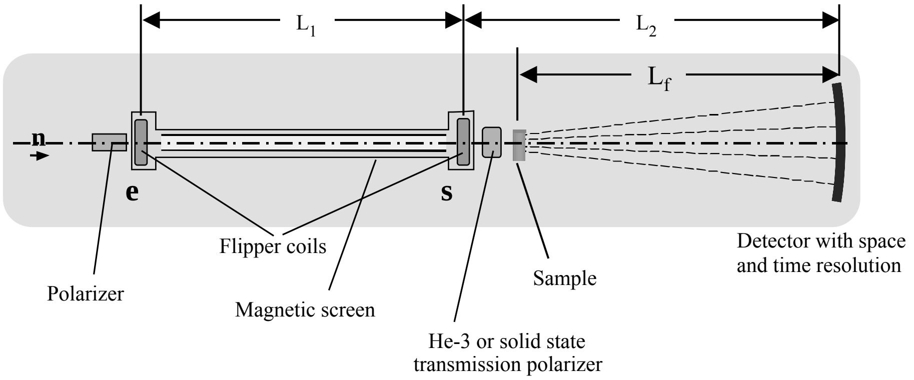

Figure 4. Schematic of the small angle neutron scattering plus modulation of intensity for zero effort (SANS + MIEZE) instrument. The beam is polarized and analyzed using ^{3}He or solid-state supermirror transmission polarizers. The entrance (e) and sample (s) rf coils operate at different frequencies ω_e and ω_s with a difference ω_d of up to 1 MHz to introduce a rapid sinusoidal oscillation of the polarization pattern. This produces an oscillation of intensity. The spin echo signal of a MIEZE instrument is the measured contrast loss of these oscillations. Typical parameters are $L_1 = 9\,\mathrm{m}$, $L_2 = 11.5\,\mathrm{m}$, and $L_f = 10\,\mathrm{m}$.

More recently, an alternative to the NRSE technique for coding the scattering angle for each neutron in a diffraction experiment has been proposed by groups in the Netherlands and the US [45,46]. The new method, which uses thin magnetic films either to define the borders of NSE precession fields or as the precession fields themselves, has been applied both to small angle-scattering and reflectometry. In SANS experiments, it has successfully extended the measurable length scales to several microns [47].

A promising variation of NRSE can be realized with two flippers driven at different frequencies. This technique is called modulation of intensity for zero effort (MIEZE) of the sample. Here the intensity at the detector is modulated with a frequency up to several megahertz. Any quasi-elastic scattering at the sample leads to a decrease of contrast of the time pattern at a thin planar detector. If this is operated with microsecond time resolution, sub-nev energy transfers during scattering are expected to be visible.

Although no MIEZE spectrometer has yet been built, the principle has been verified in Munich and a first measurement has been performed in Saclay on the modified NRSE spectrometer MUSES. Technically, it is feasible to adopt MIEZE/SANS to future pulsed sources. The only parameter depending on neutron velocity is the amplitude of the RF fields which is proportional to the neutron velocity and can easily be controlled by arbitrary function generators, already in use now at all NRSE machines. It is expected that spectrometers of the MIEZE type will be useful for observing the slow dynamics of materials such as polymers, gels, liquid crystals, and biomolecules, which require not only beam correlation times in the high nanosecond-to-microsecond range but also high lateral beam correlation lengths (10–100 nm or more). Such conditions can be met by highly collimated cold neutron beams typically found on small-angle neutron-scattering (SANS) instruments. It therefore seems attractive to insert the spin echo option in an existing long baseline SANS instrument.

2.1 Polarized samples for neutron scattering

Even though polarized neutrons can be used to separate coherent and incoherent scattering, it is sometimes useful to enhance the difference between these two types of scattering processes by polarizing the nuclei of the scattering sample. This method has proven particularly useful in biological samples, which invariably contain large numbers of protons that usually scatter neutrons incoherently. Not only does this incoherent scattering appear as a background in diffraction experiments with biological samples but also the weakness of coherent scattering by protons results in low intensity of the Bragg peaks being measured. Although replacing hydrogen with deuterium in the sample can sometimes alleviate this problem, this is often not possible because deuterated versions of many organic (particularly biological) molecules are not available. When hydrogen/deuterium substitution is impossible, dynamical pumping of the proton polarization in the sample can be used to reduce the magnitude of the incoherent scattering and increase the coherent scattering, improving the signal and reducing the noise of a diffraction measurement. Although not necessarily related to the use of polarized neutrons, this polarized sample technology is worth considering in conjunction with polarized neutrons because it provides a way of changing the relative amounts of neutron spin-flip and non-spin-flip scattering from the sample and thus an additional method for separating signals from competing effects within the sample. In the past, the technology has not found widespread use outside of Europe because of the technical overhead involved in its implementation. However, it could make a significant difference to the impact of neutrons in biological problems by allowing neutrons to complement synchrotron radiation and provide information about the all-important hydrogen positions in biological macromolecules. The method also has the potential to enhance the scattering from specific protons by selective pumping.

2.2 Polarized pulsed neutrons for fundamental neutron physics

In addition to neutron scattering, essentially all modern, intense neutron sources have had, as a component of their research programme, studies in what is now referred to as "Fundamental Neutron Physics." This field, which includes measurements of fundamental constants, precise tests of basic symmetries in particle physics, and measurements of important astrophysical and cosmological quantities, has particularly flourished through the use of intense beams of polarized neutrons from high-flux reactors (see for example [48]).

While the *sine quo non* of modern cold neutron experiments is high flux, intensity alone does not ensure a successful experiment. The sensitivity of precision measurements to systematic effects implies that one is often willing to compromise on flux to reduce systematic errors. Selection of the appropriate neutron source for a particular experiment is driven by the need to achieve the optimal balance between systematic and statistical errors. Recently, it has become clear that the pulsed nature of a spallation source offers an important opportunity for the reduction of systematic effects in many of the important fundamental neutron physics experiments (see for example [49]). The high intensities offered by next-generation sources such as the SNS will provide statistical sensitivity at a level that offers outstanding discovery potential. A key to the success of future experiments will be further refinement of the ^{3}He polarization technology.

Most modern polarized cold neutron experiments have used magnetic "supermirror" reflection devices to spin-polarize the neutron beam. These devices are simple to use and can provide quite high polarizations that exceed 99% in optimal situations. However, most fundamental physics experiments require polarization of a large cross-section beam with a broad velocity spectrum and a significant divergence. For such beams, it has proven difficult to obtain a reliable, high-accuracy measurement of the average neutron polarization to much better than about 1%. This is because the polarization from a supermirror device varies across the beam and depends on both the incident neutron direction and the velocity of the neutron. The problem is exacerbated by the fact that, for decay experiments, the appropriate polarization average must be weighted by $1/v$ to appropriately account for the probability of decay. Most polarization measurements count neutrons with an efficiency that is nearly velocity independent.

A different approach to neutron polarization relies on the use of nuclear spin polarized ^{3}He gas cells as a neutron spin filter. The neutron polarization following transmission through a polarized ^{3}He cell is given by

$$P_n(v) = \tanh\left(P_3 N l \sigma(v)\right)$$

where $P_n(v)$ is the neutron polarization, P_3 is the ^{3}He polarization, N is the ^{3}He number density in the cell, l is the helium cell thickness, and $\sigma(v)$ is the unpolarized capture cross section at neutron velocity v.

Since it is difficult to measure all of the quantities accurately within the previous equation's brackets, an accurate, *ab initio* determination of the neutron polarization is not feasible. However, by exploiting the simple and well-understood interaction between neutrons and ^{3}He at low energy, the neutron polarization can be determined accurately from the measurement of different, experimentally accessible, quantities [50,51]. At a pulsed source, the rather long time of flight (TOF) for cold neutrons (tens of milliseconds from source to apparatus at a typical installation) allows high-velocity dispersion, making it possible to relate the neutron polarization to the TOF in a remarkably simple way. We note that the strong dependence of neutron polarization on velocity means that it is much more awkward to determine the polarization of a "white beam" from a reactor [50]. The capture cross section on ^{3}He accurately follows the "$1/v$ law" with $\sigma(v) = \sigma(v_0)(v_0/v)$. Substituting into the preceding result for neutron polarization we have the following:

$$P_n = \tanh\left(\frac{P_3 N \sigma(v_0) v_0}{L} t\right) = \tanh\left(t/\tau\right)$$

where L is the distance from the source to the experiment and t is the TOF. In the preceding equation, τ is the single instrumental parameter that needs to be determined to extract an accurate polarization. For example, in the neutron-spin/beta-momentum correlation experiment, the measured asymmetry $A_{\mathrm{exp}} = AP_n$. Thus the fundamental asymmetry will have the same well-understood parametric dependence on TOF and can be extracted by a single parameter fit to the asymmetry data. In essence, this procedure provides an *in situ* determination of the polarization of the neutrons that actually undergo decay in the apparatus. We note that the ^{3}He technique provides a number of other highly redundant checks on the polarization that are not available with other schemes.

The ^{3}He technology has another substantial advantage for precision measurements. In all previous cold neutron asymmetry experiments, the neutron spin has been modulated only between "spin-up" and "spin-down." A pulsed source experiment using ^{3}He thus provides a highly accurate modulation of not only the spin direction but also the magnitude of the polarization during each pulse. This amplitude modulation offers a potentially powerful tool for the identification and elimination of systematic effects that is not possible at a continuous source.

The current level of maturity of the ^{3}He polarization technology makes it useful for the next generation of fundamental physics experiments. Nonetheless, further development will be quite valuable. Like neutron-scattering experiments, fundamental physics experiments would benefit from higher ^{3}He cell polarizations. Because these experiments can often use large cross-section beams, increasing cell size will be quite important.

3. Polarized neutron technology

Realizing the scientific advantages offered by polarized neutrons, both for neutron scattering and for fundamental physics, requires a set of technologies that have been developed over the past several decades and that are continuing to evolve. All of these technologies are now sufficiently mature to allow their use at steady-state neutron sources, albeit sometimes under conditions that are not optimal. On the other hand, these technologies are not all mature at pulsed neutron sources, and few experiments at these sources use polarized neutrons. In fact, there is only one widespread use of polarized neutrons for scattering experiments at pulsed sources: neutron reflectometry.

Two types of component essential for all polarized neutron experiments are:

1. Polarizers (or analyzers) that select a particular neutron spin state (or, equivalently, a particular direction for the neutron magnetic moment).
2. Devices to control the orientation of a neutron spin in a particular spatial direction. The simplest such device is a magnetic guide field that maintains the neutron spin parallel to an applied field. In this class of devices we also include flippers (that invert the direction of the neutron spin), $\pi/2$ spin rotators, and more complex configurations (often required for GPA) that allow the neutron spin to nutate to a chosen direction.

At steady-state neutron sources, one finds three different types of neutron polarizers: (1) magnetic mirrors or supermirrors, (2) magnetized crystals, and (3) spin filters. Each of these has its own advantages and disadvantages, and the particular solution chosen depends on the application. Broadly speaking, it is true to say that one or other of these technologies can provide what is needed for almost any polarized neutron experiment at a steady-state neutron source. For pulsed sources, however, the situation is different. First, most spectrometers at pulsed sources rely on TOF rather than monochromatization to determine neutron energies and thus use neutrons with a broad range of energies. For this reason, with very limited exceptions, crystals that prepare (monochromatic) polarized neutron beams by Bragg diffraction are not useful as beam polarizers at pulsed sources.

Supermirrors, on the other hand, have been used successfully to polarize neutron beams at pulsed sources. Used individually they produce polarized beams that are spatially narrow and of limited divergence (especially at short neutron wavelengths). Beams with these properties are suitable for PNR and have allowed this technique to be developed at pulsed neutron sources, perhaps more rapidly than it was at steady-state sources. When supermirrors are incorporated in specially designed neutron guides [52], they can overcome the beam-size and divergence limitations of single mirrors and can be adequate as polarizers for many applications at pulsed sources that use cold and thermal neutrons. They will not, however, be adequate as polarizers for hot (short-wavelength) neutrons. Such neutrons are produced copiously at pulsed spallation sources and provide a capability that is unique to this type of source.

In addition to using a broad band of neutron wavelengths, pulsed sources achieve much of their impressive performance by collecting neutrons scattered over a large angular range. Analyzing the polarization of such a divergent neutron beam is a problem that has not yet been solved, although polarized ^{3}He filters are beginning to change this. These filters have the added advantage of being able to polarize neutrons of all wavelengths without the significant practical limitations of beam size. Clearly, the development of this technology will be a key enabler for the use of polarized neutrons at pulsed spallation sources.

The magnetic guide fields used at pulsed sources to maintain neutron polarization are not significantly different from those used at steady-state sources, although somewhat more attention needs to be paid to the magnitude of the fields and the rates at which their orientations change when short wavelength neutrons are used. Spin rotators and spin flippers are, however, a different story. Although several different types of flippers have been tried at pulsed sources, most of them still require further development. Devices that rotate neutron spins through a particular angle (needed for implementation of GPA) have not yet been developed for pulsed neutron beams with broad wavelength distributions.

In any facility where polarized neutrons are to be used it will be important to ensure that the magnetic environment is well understood and controlled.

It is worth pointing out that much of the R&D activity described subsequently will require the use of a neutron beam line that can be easily reconfigured for various tests. It is important that such a beam line be available on short notice.

In the following paragraphs, we summarize the current state of several polarized neutron technologies and outline what needs to be done to improve them to the point where they can be used at pulsed neutron sources.

3.1 Development of polarized ^{3}He spin filters

The range of scientific questions that can be addressed today with polarized neutrons is restricted by the technical limitations of current neutron polarizers and analyzers. This is especially true at pulsed neutron sources. Both pulsed and continuous neutron sources need polarization analyzers that can accept highly divergent beams. Whereas reactor-based instruments can employ monochromating polarizing crystals such as Heusler alloy, polarization analysis on TOF instruments at pulsed sources often requires broadband polarizers that can operate throughout the cold, thermal, and hot neutron energy ranges. Spin filters based on the large spin dependence of the cross section for absorption of neutrons by ^{3}He gas can address these issues and have additional features. In the field of fundamental neutron physics, ^{3}He spin filters have several advantages, including low background, broadband capability, uniform polarization throughout the neutron beam, and the ability to flip the neutron polarization through spin reversal of ^{3}He spins using the adiabatic fast passage technique. Coupled with the TOF analysis intrinsic to pulsed sources, ^{3}He spin filters can be used to measure polarization with unprecedented accuracy.

For these reasons, polarized ^{3}He spin-filters were identified at the Pulsed Polarized Neutrons Workshop as a key area for development and application. ^{3}He spin-filters have already made an impact on neutron scattering at ILL [53], and an even larger impact is expected at pulsed sources. In the US, pilot experiments at the NIST Center for Neutron Research (NCNR) and intense pulsed neutron source (IPNS) in SANS [54] and reflectometry [55,56] have been performed using ^{3}He-based neutron spin filters. A spin filter is under development for a fundamental neutron physics experiment at Los Alamos Neutron Scattering Center (LANSCE) also [57]. Both the development and application of ^{3}He spin filters are critical needs to make the largest range of polarized neutron research possible at new pulsed sources being built in the US and Japan.

Two optical pumping methods have been employed to polarize the ^{3}He gas for spin filters: (1) spin-exchange (SEOP) [58], in which the gas is polarized directly at high pressure (1–10 bar), and (2) metastable exchange (MEOP) [59], in which the gas is polarized at low pressure (1 mbar) and then compressed. In the MEOP method, metastable atoms produced by an electrical discharge are optically pumped by a laser, while for the SEOP method a laser optically pumps rubidium vapor that is produced by heating the cell. The current maximum ^{3}He polarization achievable for either method is 75% for practical spin filters [60–62];

60–70% has been typical for recent applications. A spin filter with 60% ^{3}He polarization will produce 90% neutron polarization with 20% absolute transmission of neutrons [63]. (For 100% ^{3}He polarization, one would obtain 100% neutron polarization and 50% transmission, i.e., all the neutrons of one spin state would be transmitted and all the neutrons of the other spin state absorbed.) Thus for less than 100% ^{3}He polarization, there is a tradeoff between neutron polarization and neutron transmission. The product of the ^{3}He pressure and cell length can be chosen to give priority to either the polarization or the transmission. Because of the wavelength dependence of the absorption cross section, the polarization varies with neutron wavelength. Typical spin filter "thicknesses" are 7 bar cm of gas for cold neutrons (wavelength of 0.5 nm) or 20 bar cm of gas for thermal neutrons (wavelength of 0.18 nm).

The maximum value of the polarization attainable with each method is only a small piece of the story in terms of the development needed for successful use of ^{3}He spin filters. There are issues in both methods for actually obtaining and maintaining this polarization in usable spin filters. The best results for the MEOP method have been obtained with large, expensive, complex compressors [53,60], resulting in the use of transportable cells that slowly lose polarization on the beam line, rather than continuously operated devices. Compact compressors have been pursued but are not yet competitive [63–65]. Polarization storage times as long as 200 h have been obtained [67], but there are several constraints that are discussed below. Producing high polarization and obtaining long relaxation times for the demanding application of large-volume polarization analyzers remains an issue. In contrast, the SEOP method is compact and can be much more easily operated continuously on a beam line [68,69], but long relaxation-time cells and long preparation times before the experiment are still required because of the slow polarizing rate of this method. For transportable cells, optical pumping times of a few days for SEOP and less than one hour for MEOP are required before the experiment to reach the maximum polarization. In addition, large volume cells have significant demands for laser power [61,70]. The SEOP method can also be used in the MEOP "filling station" approach. Hence, for both of these methods, further development is required to actually make the large variety of spin filters that will be required for pulsed source instruments. In addition, ^{3}He spin filters require low magnetic field gradients ($(1/B_0)\times(\partial B/\partial r) < 3x10^{-4}$ per cm) [53,55], which is in direct conflict with experiments that require high field superconducting magnets to magnetize a scattering sample, for example. The technology to shield such fields, which typically involves a combination of passive mu-metal and superconducting shields [53], needs to be developed for several applications. It may also be worth considering the use of larger filter holding fields than have been employed to date.

Despite the development needs just noted, a number of applications can be pursued with the current technology of ^{3}He spin filters. Indeed, substantial work has been conducted at ILL, and tests are being conducted by the National Institute of Standards and Technology (NIST) and Indiana University at the NCNR and IPNS. For a variety of reasons, it is important that such experiments be pursued in parallel with technology development. First, such efforts will establish a base of experience at current neutron facilities that will provide critical guidance for implementation on future pulsed-source instruments. Second, it is important to note that although polarized ^{3}He technology is well developed in other fields such as nuclear physics, there are few materials scientists who have experience with the application of ^{3}He spin filters to neutron scattering. Hence, experiments are required to provide experience for neutron scatterers, as well as to reveal issues in the application of these devices. In particular, the wavelength dependence of ^{3}He spin filters will present new issues that have not been addressed in connection with the use of neutron optical polarizers. In addition, for the cases where it may be difficult to establish *a priori* whether ^{3}He spin filters or neutron optical devices are the best device, experience will provide guidance. Finally, necessity is still the mother of invention; hence, the needs of real experiments will result in increased development. A notable example of this is the LANSCE "n-p-d-gamma" experiment [57,69], which helped motivate the development of large-area, long-lifetime SEOP cells by the NIST group [67] and the on-going implementation of these cells by the University of Michigan and others for a reliable, continuously operating spin filter at LANSCE.

With this introduction, we list the following directions for R&D that emerged during the Pulsed Polarized Neutrons Workshop:

1. *Experiments at existing neutron facilities with current neutron spin filters should be pursued*. The priority should be for experiments and instruments in which existing ^{3}He spin filters can have an immediate impact and can be implemented efficiently. Currently, the only US laboratory with on-site apparatus and personnel for neutron scattering experiments with spin filters is the NCNR. Outside the US, there is a well-developed programme at ILL [53], and capabilities are developing at ISIS [71,72], Hahn-Meitner-Institute (HMI) [73], and Forschungszentrum Jülich (FZJ) [74]. In addition, there has been notable progress at the High Energy Accelerator Research Organization (KEK) in Japan [75]. A polarizing apparatus that is to be set up at the IPNS is currently under construction at Indiana University. Expertise in polarized ^{3}He exists at Los Alamos National Laboratory (LANL) [69], but so far there has not been an active programme in the application to neutron scattering. There is no activity in polarized ^{3}He at Oak Ridge National Laboratory's (ORNL's) High Flux Isotope Reactor (HFIR). Active programmes in the application of the current generation of neutron spin filters need to be cultivated at as many current neutron laboratories as possible. This goal requires not just the polarized gas apparatus and relevant personnel but also an active effort on the part of interested instrument scientists and neutron scatterers to develop applications of the technology.
2. *Despite the evolving state of spin filter technology, definition and coordination of the spin filter requirements of SNS, as well as other neutron laboratories, should be initiated*. Because there can be large differences in technical focus depending on the optical pumping method (the choice of on-line operation versus transportable cells, magnetic environment issues, etc.), it is essential that applications provide direction. The emphasis should depend on choosing the approach (or approaches) that will most efficiently address the needs of existing and planned instruments for which ^{3}He spin filters are the most practical.
3. *Whereas individual facilities may focus on one of the two optical pumping methods, development of both methods should continue, not only because of their complementary nature but also because ongoing developments in each make a single choice premature*. To date, essentially all development and application of the SEOP method has been in the US, as part of past and present application of this method for electron scattering and fundamental neutron physics. In contrast, the European community has almost exclusively focused on the MEOP method and is the leader in this area. The MEOP method has also been developed in the US in two parallel efforts: (1) construction of a large-scale compression system, similar to the European apparatus, at Indiana University [76] and (2) development of a compact compressor at the NCNR [64]. The Indiana system has not reached the performance level of the European system [77]. The compact NCNR system, which was meant as a test of the concept of a dramatically smaller, simpler, and less expensive compression method, has shown promise but is not yet competitive with the either the European system or SEOP results. Because SEOP and MEOP have been developed to a high level on opposite sides of the Atlantic, it may make sense to pursue future developments in a partnership, with US and European researchers focusing on different technologies but sharing their results.

4. *For SEOP, priorities include*:
   a. *Cell development*. Because of the long-time constants for SEOP, cells with polarized gas relaxation times of 100 hours or greater are required so that the highest polarization values can be achieved. In applications in which the gas is polarized off-line and used on the beam line, the longest possible relaxation times are particularly important. To date, such long relaxation times have been obtained with reasonable reproducibility only in carefully prepared blown glass cells made by a few groups. (The theoretical maximum of 800 h for a cell at a pressure of one bar has been approached [67]). The rubidium that is introduced for optical pumping also plays a key role in suppressing wall relaxation [78]. For future applications, flat-windowed cells (which cannot be completely blown) will be desirable for uniform thickness spin filters. Experience with such cells is even more limited, with the only development for neutron applications being pursued at NCNR and ILL. Both the achievable relaxation times and the reproducibility of obtaining these times are inferior in these cells. Studies of different processing methods and/or coatings are needed. In addition, there are construction issues with the range of cell geometries and sizes that will be needed in the future. For this latter issue, direction should be established by the needs of spectrometers that can profitably use ^{3}He spin filter technology.

      Whereas alkali-coated glass is currently the best material for cells and thus should be the first line of further development, other options should be investigated. For example, work is currently being done at the University of Virginia [79] with cells that are first coated with sol-gel [80] before the alkali is introduced. Sapphire has desirable neutron properties, but there are issues with construction of large-scale cells and with birefringence. At ILL, cells have made from single crystal silicon. Although certain pure metals, such as titanium or aluminum, can exhibit long relaxation times, no one has actually constructed cells from such materials. Recently it has also been observed that cells can become magnetized in strong magnetic fields and exhibit magnetic hysteresis [81]. Even cells that have never been in strong fields can show induced and remnant magnetization that leads to a dependence of relaxation time on magnetic field direction (or equivalently cell orientation in a given field), field magnitude, and magnetic history [82]. These effects need to be better understood.
   b. *Spectrally narrowed laser development*. Large cells require substantial amounts of laser power. A scheme has recently been developed at the University of Wisconsin to spectrally narrow the broadband commercial diode bars typically used for SEOP [83]. However, these lasers do not have the convenience of commercial fibre-coupled lasers. Further investigation is required to fully explore the utility of spectrally narrowed lasers and make them more convenient for on-line spin filter applications.
   c. *Investigation of a filling station approach*. Although a major advantage of the SEOP method is the capability to continuously optically pump on a neutron beam line, for some applications a “filling station” approach may be desirable. In this approach, one decouples the optical pumping requirements from the spin filter requirements. For example, SEOP is more efficient for small, high-pressure cells that have a favourable geometry for optical access, whereas spin filter cells may be large, low-pressure cells with special shapes for large solid-angle polarization analysis. In this case, a SEOP filling station could provide the technical simplicity of the SEOP method along with the capability to fill a large range of cells, which is currently practiced at ILL using the MEOP method.
   d. *Investigation of optical pumping of other alkalis and alkali mixtures*. It has recently been shown by collaboration between the University of Wisconsin and Amersham

    Health that optical pumping of a rubidium–potassium mixture can increase the polarizing rate of SEOP [84]. This approach could increase the range of cell relaxation times that are acceptable and thus also increase the range of tolerable magnetic field gradients. In addition, it would increase the convenience and versatility of the SEOP method by shortening the time required to polarize a cell.
    e. *Investigation of the fundamental mechanisms limiting the polarization*. It has recently been discovered at the University of Wisconsin that the polarization achievable by the SEOP method is limited to about 75% by an unknown form of relaxation that scales with the rubidium density [85]. If this relaxation could be identified and eliminated, ^{3}He polarizations approaching 100% could be possible.
5. *For MEOP, priorities include*:
    a. Establishing in the US a state-of-the-art compression apparatus comparable in performance to those at ILL and Mainz, Germany.
    b. *Cell development*. Long relaxation times are important for the MEOP method, but for somewhat different reasons than those for the SEOP method. The MEOP method has a higher polarizing rate, which lessens the need for long relaxation time cells to reach high polarizations. However, the physical scale of current compressors requires a filling station approach; hence, cells are not optically pumped on the neutron beam line. The issues for MEOP cells are similar to SEOP cells but with a few differences. MEOP cells are valved rather than sealed, which introduces additional issues in maintaining cleanliness. MEOP cells can be made of fused silica, while SEOP cell use GE180 [86] (a boron-free aluminosilicate glass) because of excessive permeation of ^{3}He at high temperatures for fused silica.
    c. *Investigation of improved compact, continuously operating compressors*. Although some work has been done in this area in both the US and Europe, a more substantial effort will be required to determine whether the highest polarizations are possible with a compact system. This is a more risky endeavour relative to either the SEOP method or the already-developed large-scale compressors. However, success would allow for the polarization rate available from the MEOP method in a system that could operate continuously on a beam line.
    d. *Investigation of the fundamental mechanisms limiting the polarization*. In the newest ILL system, there is almost no loss of polarization during compression, which now turns attention to a more precise understanding of the limits of the polarization that can be produced by MEOP. We note that there is essentially no activity in the US in fundamental studies of MEOP. Increased attention to these issues from the atomic physics community would be ideal.
    e. *Polarized gas transfer methods*. In some instruments, it may be difficult to access the ^{3}He cell. In such cases, it might be desirable to have a cell fixed in the instrument that is filled through a gas line. This has recently been successfully demonstrated on the D17 reflectometer at ILL.
6. *Development of magnetic shielding methods*. In this area, no single approach is appropriate to address all issues. The solution needs to be matched to the application, which implies that more specific knowledge of the desired applications must lead the way. For relatively modest stray fields, passive magnetic shields will be adequate. A more formidable stray field will require a combination of mu-metal and Meissner shields. In this area, ILL is clearly the leader. Recently, it has been shown that glass spin filter cells can become magnetized in strong fields, resulting in decreased relaxation time [81].
7. *Development of accurate neutron polarimetry using ^{3}He spin filters*. This area is of most importance for fundamental studies of the weak interaction with neutrons [61].

Measurements of correlations coefficient in neutron beta-decay require knowledge of the neutron polarization at the 0.1% level. This is possible using ^{3}He spin filters because the polarization can be determined by neutron transmission measurements and because the dependence of the absorption cross-section on neutron energy is well known. Development of this method would facilitate a class of experiments on fundamental physics beam lines at pulsed sources.

8. *Commercial ^{3}He gas polarizers*. These polarizers are being developed for polarized gas magnetic resonance imaging (MRI). These devices cannot be used directly as neutron spin filters because the technical needs for MRI imaging are different from those of spin filters. However, some of the R&D for the polarized gas MRI is likely to be useful for neutron spin filters. The neutron community should make use of the relevant technology from the polarized gas MRI enterprise. It is important to note that such development will require input from a broad range of researchers, including neutron scatterers, nuclear and atomic physicists, and engineers, as well as cross-disciplinary collaboration. Fostering such interaction should be a priority.

3.2 Neutron spin flippers

All instruments that use polarized neutrons at pulsed neutron sources need one or more neutron spin-flippers. To be suitable for a pulsed neutron source, the flipper either needs to function intrinsically over a broad neutron bandwidth (example: Cryoflipper) or it must be possible to change the flipping conditions according to a dynamic TOF mode (e.g. Mezei flipper with ramped currents). The kind of spin-flipper that is best suited to a particular application depends on the wavelength range to be handled, the magnetic environment, the beam size and angular coverage and space restrictions.

The current needs and priorities for the development of neutron spin-flippers at pulsed sources are:

a. Development of broad wavelength-band spin-flippers
    - push λ_{min} to shorter wavelength (a suitable goal might be 0.4 Å for diffraction instruments, 0.1 Å for high-energy inelastic spectrometers)
    - develop spin flippers that provide substantial angular coverage
    - develop spin flippers that are insensitive to magnetic field environments
    - develop spin flippers that do not pollute the magnetic environment (the latter applies particularly to RF flippers)

b. Development of computational methods
    - calculate neutron beam polarization for realistic flipper/magnetic field environments
    - integrate realistic magnetic field profiles in Monte Carlo simulation programs

Below we present a number of spin-flipper concepts that need to be evaluated, adapted, and refined for the applications at future pulsed sources. For each spin-flipper, some comments are made about the current state of the art and what advances will be necessary to ensure stable and reliable operation of these devices in pulsed-source instruments. A good summary with additional details on some types of neutron spin-flippers can also be found in reference [87]. In the end of each section, we specify the actions we judge necessary to achieve the required advances.

3.2.1 Drabkin non-adiabatic flipper. The concept of non-adiabatic coil flippers was proposed by Hughes and Burgy [88] in the early 1950’s. Appropriate field configurations achieved by mounting two dc coils with opposite polarity coaxially along the neutron beam were used by Drabkin *et al.* [89] in the 1960's and by Jones and Williams [90] in the 1970's to spin-flip neutron beams. At the flipper entrance, the field is parallel to the momentum of the neutron. This longitudinal field, which defines the quantization axis for the neutron spin, reverses its direction over a very short distance at the midpoint of the flipper. The neutron spin cannot follow this fast field change and is therefore reversed relative to the guide field direction at the exit of the flipper. Besides the coaxial configurations, other geometries were realized to achieve similar results, like the coplanar rectangular coils configuration transverse to the beam proposed by Korneev [91].

A non-adiabatic coil flipper is a relatively simple solution that requires almost no further development and adaptation. In particular, there are no special requirements for magnetic field homogeneity and stability. This kind of flipper works well for epithermal and thermal wavelengths but is usually less efficient for cold neutrons. The upper wavelength bound is typically around 10 Å, but magnetic stray fields could lower this value. Efficiencies of $> 99.5\%$ have been demonstrated for a beam diameter less than 30 mm. This configuration is currently operating in spallation sources (e.g. POSY I at IPNS). It requires space on the order of 500 mm (length) along a beam line but does not require material in the neutron beam. This design could be turned into a $\pi/2$ flipper by adding one more field component in the transition region, although its performance might not be satisfactory.

3.2.2 Mezei flipper. Initially proposed by Mezei [10], a precession coil spin-flipper is constructed using two coils with perpendicular windings that create a sharp change from a transverse guide field direction to a transverse perpendicular field direction in a well-defined region in space. The neutron spin enters and exits the coils non-adiabatically (i.e. without changing its direction). The outer coil is used to cancel the guide field that is present at the flipper location. Within the inner coil (which provides a transverse flip field perpendicular to the guide field), the neutron polarization vector precesses around the flipping field. This flipper is widely used in reactors. To make it work in spallation source instruments requires the (inner) precession coil to be operated in a pulsed current mode (i.e. with a certain waveform and with the source frequency). The Mezei flipper might be the best option for instruments with tight space restrictions (typical Mezei flippers are about 5 – 10 mm thick). This flipper can also work as a $\pi/2$ flipper when the current in the precession coil is half that required for complete spin flip.

Disadvantages of this kind of flipper include that it requires the guide field compensation coil to be finely retuned whenever residual magnetic fields change at the location of the flipper (e.g. when switching on a sample magnet) and that the procedure of determining the appropriate flip current/wavelength relationship initially could be time consuming. Also, the Mezei flipper requires having material in the beam (the wires of the coils). This could cause additional scattering effects and background signal in the detectors. Several groups are working on designs that would minimize this effect.

Tests of a prototype Mezei flipper operated in TOF mode were carried out several years ago by Fitzsimmons at LANL and more recently by SNS staff at IPNS in collaboration with Te Velthius (MSD-ANL). The electronics required to operate such a flipper in a spallation source are readily available. Good performance has been achieved for neutrons with wavelengths up to 10 Å (measurement was intensity limited). At IN15 in TOF mode, neutrons with wavelengths of 18 Å have been flipped with high efficiency using a Mezei flipper.

3.2.3 Current sheet and superconducting sheet flippers. The operation of a current sheet flipper (also called a Dabbs foil flipper [92]) is based on a non-adiabatic magnetic field transition created by a current passing through a foil and several different designs applying this concept have been suggested in the past [90,93,94]. Such foils typically operate at cryogenic temperatures to achieve higher currents and, consequently, a stronger field magnitude. A similar non-adiabatic transition between fields on both sides of the flipper foil can be achieved by using the Meissner screening effect of superconducting sheets. Niobium sheets have been in operation at ILL for many years. Recently, Fitzsimmons *et al.* [95] also used high T_C films for this purpose.

The use of current sheet flippers requires insertion of material in the beam and could be somewhat sensitive to external magnetic fields (e.g. from a sample magnet in the case of current sheets). Furthermore, the arrangement of the guide field before and after the flipper might not be straightforward. Nevertheless, this type of neutron spin-flipper works well and very stably for white neutron beams, requiring no tuning.

3.2.4 RF-gradient flipper. An RF-gradient flipper [96,97] is an adiabatic neutron spin-flipper and works with two basic magnetic fields: (1) a constant guide field with a spatial gradient along the neutron beam line and (2) an RF oscillating field. This design flips all neutrons with wavelengths larger than a certain minimum wavelength for which it is designed. There is no need for a pulsed current to be synchronized with the source. It can also be designed to work well in a variable field environment, without the need of compensation [98,99]. Another advantage of this flipper is that it does not require placing material in the neutron beam. For these reasons, the RF-gradient flipper may be the preferred flipper for most pulsed neutron applications.

Satisfactory solutions for the RF electronics are in use at several places in Russia, at the Hahn-Meitner Institute (HMI) and Forschungsmess Reaktor FRM2, and at LANSCE [100]. Gradient RF flippers have been used successfully close to a high-field cryomagnet at LANSCE.

3.2.5 Flippers with multiple current foils (Drabkin spatial spin resonance). A Drabkin spatial spin-resonance flipper [101,102] functions somewhat similarly to an RF flipper. The difference is that the adiabatic spin rotation is accomplished by passing the neutron beam through an arrangement of current sheets that create a *spatially* oscillating magnetic field instead of an RF field. For TOF operation, the guide field coils and the current sheets need to be operated with ramped currents [103,104]. Depending on the number of current sheets, the flipper can be made to be wavelength selective ($\delta E/E < 1\%$).

According to calculations, a flipper of this sort should allow dynamic energy filtering at pulsed neution source [105,106]. The complete set-up would consist of a wavelength-selective magnetic resonator (the actual flipper) and a supermirror polarizer/analyzer system. Currently, several prototypes are being built and tested in Japan [107]. SNS has developed a prototype system with which preliminary tests have been made (more thorough tests can only be done with proper instrumentation in a spallation source).

3.2.6 Options for $\pi/2$ flippers. For some applications, like spin-echo spectroscopy and three-dimensional polarization analysis instruments, $\pi/2$ flippers are needed. Mezei flippers have been successfully used for this purpose at reactors, but there are also other solutions (some of which were discussed previously). Recently, magnetic thin films have been used as $\pi/2$ flippers [46], and one can imagine ways in which their use could be extended to pulsed sources by changing the direction of the magnetic field applied to the film during each neutron pulse. Japanese scientists have reported successful construction of a $\pi/2$ flipper based on a combination of a current sheet and Helmholtz coils.

3.2.7 Remanent supermirrors. Recently, so called "remanent polarizing supermirrors" have been developed by Böni *et al.* [108]. These can be operated as spin-selecting devices and could make spin-flippers dispensable for some applications. These supermirrors are designed such that spin "up" neutrons are reflected if the magnetic coating is magnetized parallel to the guide field direction. Magnetic hysteresis allows the coatings to maintain their magnetization direction even if a small field is applied in the opposite direction (reverse fields of approximately 20 G are possible). In this state, the mirror reflects spin "down" neutrons.

3.3 Neutron spin echo developments

It is clear that future research using polarized neutrons at pulsed sources will rely heavily on ^{3}He spin filters. The properties of these systems are very different from their optical counterparts, and some thought is needed to determine how best to use them for neutron-scattering experiments. What, for example, is the implication for various experimental techniques of the rather low neutron polarization currently available with high transmission spin filters? While such low polarization is probably quite adequate for separating roughly equal spin-up and spin-down signals, the situation is quite different when the signal in one channel is an order of magnitude (or more) different from that in the other channel. The optimum polarization and transmission of the spin filters will need to be established for each potential application, as will the effect of the wavelength dependence of both of these quantities.

As detailed elsewhere in this report, there are a number of instrumental methods that involve more than a straightforward assembly of the polarizer and flipper components described previously. All of these methods involve controlling Larmor precession of the neutron spin in some manner. Traditional NSE uses large volume electromagnets designed so that the precession angle of a neutron spin depends only on the neutron speed and not (in lowest order) on divergence or position within the neutron beam. NSE methods to code the angular trajectory of a neutron, on the other hand, require fields whose boundaries are inclined to the neutron beam. MIEZE and NRSE methods use both homogeneous static magnetic fields and RF fields. Finally, GPA uses precession of neutron spins to arrange for the spins of neutrons incident on and scattered from a sample to be aligned along well-defined spatial directions.

Traditional NSE has been in use at continuous neutron sources for more than two decades, although experience with the method in the US is limited to a recently constructed spectrometer at NIST. This method has recently been tested in TOF mode using the IN15 spectrometer at ILL and is likely the easiest of the Larmor precession techniques to transfer to pulsed neutron sources, once some of the problems with polarizers and flippers (including $\pi/2$ rotators) described previously have been adapted to the particular requirements (wavelength range, time structure, divergence, etc.) of pulsed neutron sources. The (energy-resolving) NRSE method has not been attempted in TOF mode, but the development necessary to ensure that this can be done is relatively straightforward since it involves only the ramping of magnetic fields during each neutron pulse. It is possible that the same solution could be applied to implementing GPA at pulsed sources. Alternatively, it might be worth exploiting the broad wavelength band available at pulsed sources to simultaneously probe different components of the susceptibility tensor (just as one uses this bandwidth, for example, to simultaneously probe different lattice spacings in a diffraction experiment). R&D will be needed to determine the optimal solution.

Finally, there are several instrumental techniques that are either new or still under development at continuous sources and that hold the promise of enhancing the resolution and sensitivity of neutron scattering. One such technique is NRSE applied to the measurement of line widths of collective excitations or to measurement of very small scattering angles. In the context of surface studies, the latter has been dubbed SERGIS (spin echo resolved grazing incidence scattering); while for scattering from bulk samples, it is known as SESANS. Although the NRSE-based SERGIS and SESANS techniques can likely be implemented at a pulsed source with about the same level of difficulty as the standard energy-resolving NRSE method, it is less obvious how measurement of phonon line shapes would be implemented using TOF methods. Significant R&D will be needed. Similarly, new techniques that use magnetic thin films for SERGIS and SESANS [45,46] will need to be explored at both continuous and pulsed sources before they can become part of the standard arsenal of neutron-scattering methods.

The following R&D activities will be required:

1. Develop SERGIS and SESANS at existing reactor-based sources to assess their potential for obtaining unique scientific information.
2. Develop the technology required (such as wavelength independent $\pi/2$ rotators) to test SERGIS at existing pulsed sources.
3. Construct a prototype SERGIS spectrometer at an existing pulsed source. The ASTERIX spectrometer at LANSCE could provide an appropriate base for such an instrument.
4. Continue to actively involve the neutron community to determine whether results achieved in prototype tests warrant construction of a dedicated instrument.

Design concepts for a MIEZE spectrometer have been developed and presented in several forums including a workshop on NSE Techniques at Pulsed Sources held at ANL in July 2002. Because MIEZE is considered technically feasible for adapting to a pulsed source, it is appropriate to begin exploring the practicalities of such a spectrometer. In particular, it will be necessary to:

1. Assess the fast detector electronics that will be needed for MIEZE.
2. Calculate the performance of MIEZE based on the performance of individual components operating in TOF mode.
3. Construct a prototype including polarizers and spin flippers and perform tests to assess the accuracy of calculations. The existing SAD small-angle scattering beam line at IPNS could be used for this purpose.

3.4 Developments in polarized-neutron optics

Many experiments in neutron scattering are limited by flux and sample size. Optical elements have been developed that enhance the neutron flux on the sample with little or no compromise to the overall instrument resolution. Two types of scattering experiments in which these limitations are particularly severe are small-angle scattering, where the samples are often large but weakly scattering, and single-crystal diffraction, where the samples are often quite small. There are possible solutions to this problem that will work for polarized beams both for SANS and diffraction. Hexapole lenses are a suitable polarization-sensitive flux-gathering optic for SANS, and Kumakov lenses are suitable for small crystals on single-crystal instruments.

3.4.1 Hexapole lens. A magnetic hexapole field acts as a lens for neutron beams, convergent for one spin component of the beam while divergent for the opposite. This device has already been demonstrated in steady-state monochromatic operation at the JRR-3 reactor in Japan by the neutron optics and detector group at RIKEN. Efforts are under way to develop a version of the device based on superconducting electromagnets. This would give the device a large enough open area to be practical as a polarization-dependent, flux-gathering optic for small-angle scattering, while at the same time allowing the field strength to be controlled in a time-dependent fashion to maintain a fixed focal length over a broad range of wavelengths for use on TOF instruments.

The following R&D will be required for further development of magnetic lenses:

1. Calculate the control parameters for a pulsed, superconducting hexapole lens for operation at pulsed sources. Develop and test the control electronics.
2. Explore the possibility of using wavelength-compensating material lenses in conjunction with the superconducting lens to extend its wavelength range.
3. Design and construct a prototype lens element to test performance in pulsed-beam operation. These tests will require a polarized beam, a high-resolution area detector, and polarization analysis capability. Small-angle scattering beam lines at either IPNS or LANSCE could be used for these tests

3.4.2 Magnetic Kumakov lens. In the past decade, the Kumakov (focusing capillary, lobster-eye) lens has been demonstrated to be an effective means for boosting flux on a very small sample area. This device is a monolithic assembly of confocal tapered waveguides. These devices show great promise in the field of single-crystal diffraction. To date, all such devices have been constructed of silica and hence do not influence the polarization of the neutron beam. Thus, they can be used in conjunction with spin filters for studies of the magnetic scattering from small single crystals. However, another interesting possibility exists: the lenses themselves can be manufactured from a magnetically birefringent material (a cobalt-iron alloy, for example) so that they selectively focus one spin state of the incident beam and produce a polarized beam on the sample. To realize such a possibility R&D is needed:

1. Determine suitable materials for fabricating a polarizing Kumakov lens both from a neutron-optical and from a fabrication standpoint.
2. If suitable materials can be found, design and construct a prototype lens element and test its focusing and polarization efficiencies. The single-crystal diffractometer beamlines at IPNS or LANSCE could be used for these tests.

3.5 Software for polarized neutron instrumentation design

Workshop participants identified several areas of importance to the development of simulation software that advances polarized neutron instrumentation and techniques. Reliable and powerful simulation software and advanced neutron instrumentation development are now inseparable commodities.

3.5.1 Existing software packages available to the public. Examples of well-documented, “whole instrument” simulation software include the Monte Carlo packages recently summarized in *Neutron News*, Vol. 13, Issue 4. These include VITESS,[†] maintained by HMI, Germany, that incorporates modules for supermirror polarizers and benders, ^{3}He polarizers, simple flipper coils, precessions in inhomogeneous fields, and precessions in rotating fields (useful for RF spin flippers). Compound devices such as polarizing cavities or beam-splitter polarizers could be constructed using the available modules. Expressed future development goals for VITESS include comprehensive simulation capabilities for NSE spectrometers,

---

[†] http://www.hmi.de/projects/ess/vitess/

including TOF NSE. VITESS has some development cross-links with MCSTAS[†], supported by Risø National Laboratory, Denmark, and ILL, France. MCSTAS appears to have limited polarized neutron capability apart from ^{3}He polarizers in its current form. RESTRAX[‡] was developed jointly at the Nuclear Physics Institute, Czech Republic, and ILL. Oriented towards triple-axis instruments, RESTRAX has some capability for simulating other instrument architectures and offers the possibility to model supermirror benders.

Packages developed in the US include NISP (LANSCE/LANL)[§] and IDEAS (SNS/ORNL).[¶] NISP is at an advanced level of development and offers spin-dependent transport calculations and superposition of magnetic fields within arbitrarily chosen regions. Both spin state and spin precession are accounted for. NISP offers a well-established input data format (MCNP), a web-based graphical user interface (GUI) for geometry building, file format conversion, and 3-D instrument visualization utilities. However, NISP still lacks "plug and play" modules for specific polarization devices. It is actively encouraging contributions from the user community. IDEAS is an evolving package whose polarized neutron components currently include various types of polarizers and spin flippers. It has a standardized module interface that offers flexibility for user-contributed modules but is currently single platform (Windows). User-contributed modules are incorporated into the library subject to their proven test results and availability of documentation.

3.5.2 Development of existing software. Multi-platform packages such as VITESS and NISP and the (currently) single-platform IDEAS have well thought out program structures where the possibility of distinguishing neutron polarization states has been integrated from the outset. Such programmes feature: modular structure; standardized module interfaces; ease of use; GUI-assisted, geometry-building tools; evolving libraries of modules for common "standard" optical elements and samples; and the possibility for entry of numerical data (e.g. most packages model supermirror polarizers by allowing input of both a spin up and a spin down reflectivity curve). These features and good documentation make them ideal candidates for module development for polarized neutron devices.

These packages are not restricted to instrument design and optimization problems but also provide powerful tools for correcting and/or analyzing experimental data from existing instruments and a learning tool where simulated experiments can be performed using appropriate sample scattering models. The Monte Carlo technique is particularly powerful because quantities can be tallied that are otherwise experimentally inaccessible. However, apart from perhaps NISP, input of spin-dependent, sample-scattering laws for simulating polarization analysis experiments with magnetized samples appears to be an area where some work is needed. Other areas of need appear to include accounting for adiabaticity or non-adiabaticity of spin rotators and guide fields, assessment of polarization homogeneity at arbitrary positions in a neutron beam, and the possibility for defining complex field maps within specific regions. Such field maps can be generated by sophisticated, commercially available simulation software. There is also a ubiquitous need for more powerful graphical tools for representing geometries and simulation results. The possibility to export data into the more commonly available commercial graphics packages could be a useful first step in this area.

3.5.3 Standardization of packages. It seems neither reasonable nor straightforward to enforce a standardized input for packages that are already at an advanced stage of

---

[†] http://neutron.risoe.dk/mcstas/

[‡] http://omega.ujf.cas.cz/restrax/guide/whatis.html

[§] http://strider.lansce.lanl.gov/NISP/Welcome.html

[¶] http://www.sns.anl.gov/pdfs/montecarlo/NN_v13n4_pgs30-34.pdf development. For example, NISP is heavily founded on the MCNP format. However, module interfaces should be standardized within a given package and possibly a standard should be agreed upon for future developments. Also, input conversion utilities that allow certain problems to be run using several packages with a minimum of learning effort or setup time might be feasible.

The European network software for computer aided neutron scattering[†] (SCANS) has the following expressed mission statement: *software development that enables more cost effective and scientifically productive use of existing neutron scattering facilities in Europe*. A similar objective should be pursued in the US.

3.5.4 Benchmarking, quality assurance, and version control. Libraries of realistic and efficient models for optical elements are essential for making reliable predictions of complex instrument behaviour. These models cannot be tested without suitable experimental data. This inevitably requires cooperation from existing neutron sources with respect to ensuring availability of test beam facilities and allocation of adequate beam time for these apparently mundane purposes. Furthermore, measured data are used as the model in some simulations.

User-submitted modules must be subject to quality control and approval by the authors of packages before they are integrated into libraries. Release of source code is useful for understanding algorithms that cannot be fully documented. However, a release of unauthorized versions of the source code by individuals is often an undesirable consequence. This practice should be strongly discouraged by providing easy and rapid processing mechanisms for code (and accompanying documentation) submitted privately to package administrators and/or by facilitating temporary plug-ins of user-customized modules, where the user assumes responsibility for the reliability of the results. These concerns seem to be well addressed for some of the projects cited previously.

3.5.5 Publicity and user education. An important step in discouraging both duplication of effort and distribution of software of dubious reliability lies in continual publicity of available simulation packages and provision of education tools (on-line tutorials, workshops etc.). These constitute some of the activities of the European group SCANS, cited in Section 3. The authors of VITESS are to be commended on making available demonstration simulation problems that greatly accelerate the software learning process.

3.5.6 Attracting software developers. A point of concern is that of attracting motivated professional software developers to work on these important projects. Poorer long-term career prospects and remuneration compared to the software industry norms appear to be partially responsible. Often, research scientists assume these roles in addition to their other responsibilities.

4. Community development

Although US scientists participated in and sometimes led the early development of polarized neutron techniques (see section on historical background), the past three decades have seen a marked shift towards European dominance in this field. If polarized neutron techniques are to be implemented at existing and future pulsed neutron sources (such as the SNS and J-PARC, already under construction in the US and Japan respectively), existing worldwide expertise will need to be harnessed to develop the necessary technologies described in this report.

[†] http://www.studsvik.uu.se/software/scans/scans.htm-Projects

While this may not be too difficult in some areas (such as polarized ^{3}He) where there are already established programmes in both Europe and the US, there are other areas (such as GPA) where there are large disparities in expertise between different world regions. There are also broad areas (such as experience with building and operating polarized neutron spectrometers or designing experiments with polarized neutrons) populated only by a very few US and Japanese experts. Most of these individuals have little or no opportunity to pass on their knowledge to others in the current environment. Furthermore, the number of spectrometers equipped to use polarized neutrons is very low in the US and there are almost no facilities available in the US for testing polarizers and other essential hardware elements.

Solutions to these problems are easily devised and a list of relevant steps (see below) was produced at the workshop. The impediment to implementing these solutions is more one of political will and financial means rather than one of designing suitable solutions. Ideas that emerged at the workshop were:

- Promote collaborations between US, European and Japanese researchers in polarized neutron experiments at facilities and universities in each of these regions. A particularly useful way of sharing expertise is to station post-doctoral fellows at foreign institutions.
- Increase beam time usage on existing polarized neutron instruments in all world regions to the extent that this is feasible. Since these instruments generally operate full time in polarized mode, the only real way to increase beam time is to add operating time.
- Provide test beam lines and support facilities, particularly in the US where these are rare commodities. In spite of suggestions that have been made for such facilities in the past, little has been done to provide them. While everyone acknowledges that little publishable science will come from such installations, the world's best neutron facilities have recognized that they can only stay at the forefront by providing neutron beams for technique development. Managers of these facilities have taken the necessary steps, sometimes in the face of criticism from users who saw that these beam-lines could be used full-time for neutron scattering investigations.
- Provide training and education of new researchers and students. There is almost no formal training in the use of polarized neutrons. The subject is not taught through lecture courses nor is there much opportunity to learn it in practice, except through participation in scheduled user experiments, which are often so hectic that little knowledge transfer can take place. The short training programme being offered at NIST in connection with the 2004 PNCMI conference was a small attempt to make progress in educating users, but more needs to be done. An introductory course on polarized neutrons should be incorporated in one or more of the neutron schools held in various countries.

Acknowledgements

The workshop on which this report is based was funded by grants from the US Department of Energy, Office of Basic Energy Sciences and by the US National Science Foundation.

References

[1] J. Chadwick, *Proc. Roy. Soc.* (London) **136**, 692 (1932).
[2] F. Bloch, *Phys. Rev.* **50**, 259 (1936).
[3] O. Halpern and M. Johnson, *Phys. Rev.* **55**, 898 (1939).

[4] Y. Izyumov and S. Maleyev, *Sov. Phys. -JETP* **14**, 1668 (1962).
[5] M. Blume, *Phys. Rev.* **130**, 1670 (1963).
[6] M. Blume, *Phys. Rev.* **133**, A1366 (1964).
[7] Y. Izyumov, *Sov. Phys. -Usp.* **16**, 359 (1963).
[8] R. Schermer and M. Blume, *Phys. Rev.* **166**, 554 (1968).
[9] R. Moon, T. Riste and W. Koehler, *Phys. Rev.* **181**, 920 (1969).
[10] F. Mezei, *Z. Phys.* **255**, 146 (1972).
[11] F. Mezei, A. Dagleish, *Commun. Phys.* **2**, 41 (1977).
[12] O. Schaerpf, *Physica* **B 156–157**, 639 (1989).
[13] P. Brown and J. Forsyth, *Br. J. Phys.* **1529**, 15 (1964).
[14] A. Freund, R. Pynn, W.G. Stirling and C.M.E. Zeyen, *Physica* **120B**, 86 (1983).
[15] F. Mezei, *Physica* **86–88**, 1049 (1977).
[16] M.T. Rekveldt, *Z. Phyzik* **259**, 391 (1973).
[17] H. Alperin (Editor), *International Conference on Magnetism*, MosCow, ed., Proc. ICM-73, (1973).
[18] F. Tasset, *Physica* **B 157–157**, 627 (1989).
[19] F. Tasset, T. Chupp, J. Pique, A. Steinhof, A. Thompson, E. Wasserman and M. Ziade, *Physica*. **B 180 and 181**, 896 (1992).
[20] L. Passell and R.I. Schermer, *Phys. Rev.* **146**, 150 (1966).
[21] H. Humblot, W. Heil, E. Lelièvre-Berna and F. Tasset, *Neutron News*, edited by G. Lander (Taylor & Francis Inc., 1997), Vol. 13/No. 1, p. 27.
[22] J. Schweizer and E. Ressouche, *Magnetism: Molecules to Materials*, edited by J.S. Miller and M. Drillon (Wiley -VCH, 2001), p. 235.
[23] S. Ferlay, T. Mallah, R. Ouahès, P. Veillet and M. Verdaguer, *Nature* **378**, 701 (1995).
[24] J. Overgaard, B.B. Iversen, S.P. Palii, G.A. Timco, N.V. Gerbeleu, L. Singerean and F.K. Larsen, *Chem. Eur. J.* **8**, 2775 (2002).
[25] S. Pillet, M. Souhassou, Y. Pontillon, A. Caneschi, D. Gatteschi and C. Lecomte, *N. J. Chem.* **25**, 131–143 (2001).
[26] J. Déportes, R. Lemaire, R. Ouladdiaf, E. Roudaut and F. Sayetat, Antiferromagnetism of the Hexagonal Laves Phases $ThMn_2$, *J. Magn. Magn. Mater.* **70**, 191–193 (1987).
[27] A. Purwanto, V. Sechovsky, L. Havela, R.A. Robinson, H. Nakotte, A.C. Larson, K. Prokes, E. Bruck and F.R. deBoer, *Phys. Rev.* **B 53**, 758–765 (1996).
[28] H. Nakotte, N.R. Dilley, M.S. Torikachvili, H.N. Bordallo, M.B. Maple, S. Chang, A. Christianson, A.J. Schultz, C.F. Majkrzak and G. Shirane, *Physica* **B 261**, 280 (1999).
[29] P.G. Radaelli, R.M. Ibberson, D.N. Argyriou, H. Casalta, K.H. Andersen, S.W. Cheong and J.F. Mitchell, *Phys. Rev.* **B 63**, 172419 (2001).
[30] S. Maleyev, *Physica* **B 297**, 67 (2001).
[31] M.R. Fitzsimmons, S.D. Bader, J.A. Borchers, G.P. Felcher, J.K. Furdyna, A. Hoffmann, J.B. Kortright, Ivan K. Schuller, T.C. Schulthess, S.K. Sinha, M.F. Toney, D. Weller and S. Wolf, *J. Magn. Magn. Mater.* **271**, 103 (2004).
[32] M. Steiner, K. Katurai, J.K. Kjems, D. Petitgrand and R. Pynn, *J. Appl. Phys.* **61**, 3953 (1987).
[33] P.W. Mitchell, R.A. Cowley and R. Pynn, *J. Phys.* **C17**, L875 (1984).
[34] K. Kakurai, R. Pynn, M. Steiner and B. Dorner, *Phys. Rev. Lett.* **59**, 708 (1987).
[35] M. Steiner, K. Kakurai, W. Knop, R. Pynn and J.K. Kjems, *Solid State Commun.* **41**, 329 (1982).
[36] J.P. Boucher, L. P. Regnault, R. Pynn, J. Bouillot and R.P. Renard, *Europhys. Lett.* **1**, 415 (1986).
[37] P. Bourges, B. Keimer, L.P. Regnault and Y. Sidis, *J. Supercond.* **13**(5), 735 (2000).
[38] D.N. Argyriou, T.M. Kelley, J.F. Mitchell, R.A. Robinson, R. Osborn, S. Rozenkranz and J.D. Jorgensen, *J. Appl. Phys.* **83**, 6374 (1998).
[39] P. Brown, *Physica* **B 297**, 198 (2001).
[40] P. Brown, J. Forsyth, E. Lelièvre-Berna and F. Tasset, *J. Phys. Cond. Matter* **14**, 1 (2002).
[41] See, for example, in *Neutron Spin Echo Spectroscopy—Basics, Trends and Applications*, edited by F. Mezei, C. Pappas and T. Gutberlet (Springer-Verlag, Heidelberg, 2003).
[42] F. Mezei, *Neutron Inelastic Scattering* (IAEA, Vienna, 1978), p. 125.
[43] R. Pynn, *Neutron Spin Echo, Lecture Notes in Physics*, edited by F. Mezei (Springer, Berlin, 1980), Vol. 128, p. 159.
[44] T. Keller, R. Gaehler, H. Kune and R. Golub, *Neutron News* **6**(3), (1995).
[45] M.T. Rekveldt, *NIM* **B 114**, 366 (1996).
[46] R. Pynn, M.R. Fitzsimmons, M.T. Rekveldt, J. Major, H. Fritzsche, D. Weller and E.C. Johns, *Rev. Sci. Instrum.* **73**, 2948 (2002).
[47] T. Krouglov, W.H. Kraan, J. Plomp, M.T. Rekveldt and W.G. Bouwmann, *J. Appl. Cryst.* **36**, 816 (2003).
[48] O. Zimmer, *et al.*, *NIM* **A440**(3) 3 (2000).
[49] C.R. Gould, G.L. Greene, F. Plasil, W.M. Snow, *Fundamental Physics with Pulsed Neutron Beams* (World Scientific, 2001).
[50] G.L. Greene, A.K. Thompson and M.S. Dewey, *Nucl. Instrum Methods* **A 356**, 177 (1994).
[51] D.R. Rich, *et al.*, *NIM* **A 440**, 772 (2000).
[52] K. Al Usta, R. Gaehler, P. Boni, P. Hank, R. Kahn, M. Koppe, A. Menelle and J.W. Petry, *Physica* **B241**, 77 (1997).
[53] T.W. Roberts, *et al.*, *Physica* **B 297**, 282 and references therein (2001).

[54] T.R. Gentile, G.L. Jones, A.K. Thompson, J. Barker, C.J. Glinka, B. Hammouda, W. Lynn, *J. Appl. Crystallogr.* **33**, 771 (2000).

[55] W.C. Chen, T.R. Gentile, K.V. O'Donovan, J.A. Borchers and C.F. Majkrzak, *Rev. Sci. Instrum.* **75**, 3256 (2004).

[56] W.C. Chen, *et al.*, *Physica* **B 335**, 196 (2003) and references therin.

[57] Y. Masuda, *et al.*, *Nucl. Phys.* **A 721**, 485C (2003).

[58] M.A. Bouchiat, T.R. Carver and C.M. Varnum, *Phys. Rev. Lett.* **5**, 373 (1960).

[59] F.D. Colegrove, L.D. Schearer and G.K. Walters, *Phys. Rev.* **132**, 2561 (1963).

[60] M. Batz, *et al.*, accepted by *J. Res. Natl Inst. Stand. Technol.*

[61] T.R. Gentile, *et al.*, accepted by *J. Res. Natl Inst. Stand. Technol.*

[62] T.R. Gentile, *et al.*, accepted by *Physica* **B**.

[63] G.L. Jones, *et al.*, *Nucl. Instrum. Methods* **A 440**, 772 (2000).

[64] T.R. Gentile, D.R. Rich, A.K. Thompson, W.M. Snow and G.L. Jones, *J. Res. Natl Inst. Stand. Technol.* **106**, 709 (2001).

[65] *P.J.* Nacher, *et al.*, *Eur. Radiol.* **9**, B18 (1999).

[66] S. Fichele, Ph.D. thesis, University of Nottingham (2002).

[67] D.R. Rich, T.R. Gentile, T.B. Smith, A.K. Thompson and G.L. Jones, *Appl. Phys. Lett.* **80**, 2210 (2002).

[68] G.L. Jones, *et al.*, accepted by *Physica* **B**.

[69] S. Page, *et al.*, accepted by *J. Res. Natl Inst. Stand. Technol.*

[70] *B.* Chann, *et al.*, *J. Appl. Phys.* **94**, 6908 (2003).

[71] S. Parnell, accepted by *Physica* **B**.

[72] G.W. Wallidge, accepted by *Physica* **B**.

[73] V. Hupanu, A. Gorzel, J. Klenke and A. Rupp, *Physica* **B 335**, 205 (2003).

[74] R. Mueller, *et al.*, *Physica* **B 335**, 278 (2003).

[75] T. Ino, accepted by *Physica* **B**.

[76] C.D. Keith, *et al.*, *Nucl. Instrum. Methods* **A 402**, 236 (1998).

[77] D.S. Hussey, *et al.*, submitted to *Rev. Sci. Instrum.*

[78] W. Heil, *et al.*, *Phys. Lett.* **A 201**, 337 (1995).

[79] W.A. Tobias and G.D. Cates, private communication.

[80] M.F. Hsu, G.D. Cates, I. Kominis, I.A. Aksay and M. Dabbs, *Appl. Phys Lett.* **77**, 2069 (2000).

[81] R.E. Jacob, S.W. Morgan, B. Saam and C. Leawoods, *Phys. Rev. Lett.* **87**, 143004 (2001).

[82] R.E. Jacob, J. Teter, B. Saam, W.C. Chen, T.R. Gentile, *Phys. Rev.* **A 69**, 021401(R) (2004).

[83] B. Chann, I. Nelson and G. Walker, *Opt. Lett.* **25**, 1352 (2000).

[84] E. Babcock, *et al.*, *Phys. Rev. Lett.* **91**, 123003 (2003).

[85] B. Chann, E. Babcock, L.W. Anderson and T.G. Walker, *Phys. Rev.* **A 66**, 032703 (2002).

[86] GE Lighting Component Sales, Bldg. 315D, 1975 Noble Rd., Cleveland, OH 44117.

[87] W.G. Williams, *Polarized Neutrons (Oxford Series on Neutron Scattering in Condensed Matter No.1)* (Oxford Science Publications, 1988).

[88] D.J. Hughes, T. Burgy, *Phys. Rev.* **81**, 498 (1951).

[89] G.M. Drabkin, E.I. Zabidarov, Ya.A. Pasman and A.I. Okorokov, *Sov. Phys. JETP* **29**, 261 (1969).

[90] T.J.L. Jones and W.G. Williams, *Nucl. Instrum. Methods* **152**, 463 (1978).

[91] D.A. Korneev, *Nucl. Instrum. Methods* **169**, 65 (1980).

[92] J.W.T. Dabbs, L.D. Roberts and S. Bernstein, 1955, Oak Ridge National Laboratory, Report ORNL-CF-55-5-126.

[93] K. Abrahams, O. Steinsvoll, P.J.M. Boongaarts and P.W. de Lange, *Rev. Sci. Instrum.* **33**, 524 (1962).

[94] J.B. Forsyth, *At. Energ. Rev.* 17-2, 345 (1979).

[95] M.R. Fitzsimmons, R. Pynn, M. Luett, H. Kinder, W. Prusseit, *Physica* **B 241–243**, 121 (1997).

[96] A.N. Bazhenov, V.M. Lobashev, A.N. Pirozhkov and V.N. Slusar, *Nucl. Instr. Methods* **A 332**, 534 (1993).

[97] S.V. Grigoriev, A.I. Okorokov and V. Runov, *Nucl. Instr. Methods* **A 384**, 451 (1997).

[98] T. Keller, T. Krist, A. Danzig, U. Keiderling, F. Mezei and A. Weidenmann, *Nucl. Instr. Methods* **A 451**, 474 (2000).

[99] A.A. Parizzi, F. Klose and V. Christoph, *Physica* **B 356**, 156 (2005).

[100] M.R. Fitzsimmons and C.F. Majkrzak, Polarized neutron magnetic reflectometry. *Modern Techniques for Characterization of Magnetic Materials* (Kluer Academic Publishers, in press).

[101] G.M. Drabkin, V.A. Trunov, V. Runov, *Sov. Phys. JETP* **54**, 369 (1968).

[102] M.M. Agamaliam, G.M. Drabkin and V.I. Sbitnev, *Rev. Sect. Phys. Lett.* (Physics Reports) **168**(5), 265 (1988).

[103] G. Badurek, A. Kollmar, A. Seeger, W. Schalt, *Nucl. Inst. Methods* **A 309**, 275 (1991).

[104] A.A. Parizzi, W.-T. Lee and F. Klose, *Appl. Phys.* **A 74**, S1607 (2002).

[105] A.A. Parizzi, W.-T. Lee and F. Klose, *SPIE Conference Proceedings* (Neutron Optics) **4509**, 114 (2001).

[106] A.A. Parizzi, W.-T. Lee, G.P. Felcher and F. Klose, *J. Neutron Res. II* **1–2**, 51 (2003).

[107] M. Takeda and Y. Endoh, *Physica* **B 267–268**, 185 (1999).

[108] P. Böni, D. Clemens, M. Senthil Kumar and C. Pappas, *Physica* **B 267–268**, 320 (1999).