Polarisation analysis on the LET time-of-flight spectrometer

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Abstract. We present a design for implementing uniaxial polarisation analysis on the LET cold neutron time-of-flight spectrometer, installed on the second target station at ISIS. The polarised neutron beam is to be produced by a transmission-based supermirror polariser with the polarising mirrors arranged in a “double-V” formation. This will be followed by a Mezei-type precession coil spin flipper, selected for its small spatial requirements, as well as a permanent magnet guide field to transport the beam polarisation to the sample position. The sample area will contain a set of holding field coils, whose purpose is to produce a highly homogenous magnetic field for the wide-angle $^3$He analyser cell. To facilitate fast cell changes and reduce the risk of cell failure, we intend to separate the cell and cryostat from the vacuum of the sample tank by installing both in a vessel at atmospheric pressure. When the instrument upgrade is complete, the performance of LET is expected to be commensurate with existing and planned polarised cold neutron spectrometers at other sources. Finally, we discuss the implications of performing uniaxial polarisation analysis only, and identify quasi-elastic neutron scattering (QENS) on ionic conducting materials as an interesting area to apply the technique.

1. Introduction

Longitudinal neutron polarisation analysis grants the ability to separate the components of the neutron scattering cross section, thereby permitting the isolation of weak magnetic or structural signals, as well as giving direct access to the directional components of the sample magnetisation [1]. While the technique has been applied successfully on a wide range of instruments at both reactor and spallation sources, its extension to wide-angle time-of-flight spectrometers has proved challenging. Indeed, despite 10 years passing since the first proof of principle [2], the technique is still not in routine use. The reasons for this are manifold. Firstly, time-of-flight spectroscopy remains a flux limited technique, requiring large samples and long counting times, and it is only recently that polarisers and analysers with a sufficient transmission to permit experiments on reasonable timescales have become available. Second is the issue of analysing the scattered neutron polarisation over the large solid angles covered by the detectors on direct geometry time-of-flight spectrometers. Two options for achieving this exist: channeled supermirror-based devices and polarised $^3$He filters. For the former, acceptance of beam divergence from large samples, the outgoing divergence from the device, and shadowing of the detectors by the absorbing blades all complicate data collection and analysis. The cost of a large supermirror array can also be prohibitive. For the latter, maintaining the $^3$He polarisation over a large
Figure 1. Schematic of the LET instrument viewed vertically. Five sets of disc choppers (C1-C5) select one or more neutron wavelength (Pulse Removal – PR, Contaminant Removal – CR), shape the pulse (Pulse Shaping – PS, Resolution – R), and eliminate frame overlap (Frame Overlap – FO). The magnified regions show the positions of the polariser and flipper and the analyser, respectively. Once polarised (see Section 3.1 and Figure 2), the neutron beam can be flipped by a precession coil flipper (see Section 3.2). It is then transported to the sample by guide fields and a set of coils (see Section 3.4), before being analysed by a wide-angle $^3$He spin filter analyser (see Section 3.3). Scattering from the analyser cell and the sample environment is eliminated by a radial oscillating collimator.

The LET spectrometer is a direct geometry time-of-flight spectrometer located on the second target station at the ISIS pulsed neutron and muon source in the United Kingdom. It is installed on a coupled hydrogen moderator, which provides a neutron spectrum in the wavelength range $\lambda = 2 - 10$ Å[12]. A set of five disc choppers allow for so-called repetition rate multiplication, whereby the time-frame between neutron pulses is efficiently filled by measuring spectra at several different incident energies $E_i$ [12, 13]. The neutron flux at the sample position at a resolution of $\Delta E/E_i = 2\%$ and $\lambda = 5$ Å, corresponding to the maximum in the wavelength spectrum, is $\Phi = 4.4 \times 10^4$ n cm$^{-2}$ s$^{-1}$, close to those of CNCS at the Spallation Neutron Source [14] and IN5 at the Institut Laue-Langevin [15] for the same conditions. A unique feature of LET among cold time-of-flight spectrometers is its large detector coverage, approaching $\pi$ steradians, making it ideally suited for collecting four-dimensional $S(Q,\omega)$ maps of excitations in single crystals. Its excellent energy resolution, as low as 1% at $E_i = 1$ meV, also makes it an excellent choice for quasi-elastic neutron scattering (QENS) on materials ranging from biological systems to polymers to inorganic ion conductors. The key parameters of the instrument are summarised in Table 1.
Table 1. Key parameters for the LET spectrometer.

| Parameter          | Value                                                                 |
|--------------------|----------------------------------------------------------------------|
| $E_i$              | 0.8 – 20 meV ($\lambda = 2 – 10$ Å)                                  |
| $\Delta E/E_i$     | 0.5 – 4%                                                             |
| $\Phi$ (5 Å, 2%)   | $\Phi = 4.4 \times 10^4$ n cm$^{-2}$ s$^{-1}$ (unpolarised)          |
| Beam size          | 20 $\times$ 40 mm$^2$                                              |
| Detectors          | $^3$He PSD, $-40^\circ \leq \theta \leq 140^\circ$, $-30^\circ \leq \gamma \leq 30^\circ$ |
| Polarizer          | $m = 5$ FeSi channelled “double-V” cavity                           |
| Flipper            | ramped precession coil (Mezei)                                      |
| Analyser           | wide-angle $^3$He spin filter                                       |

We present here the design for a longitudinal polarisation analysis option on LET, the construction of which is currently in progress. We plan to overcome the difficulties highlighted above by choosing the simplest possible implementation of polarisation analysis: the beam will be polarised by a solid state super-mirror device, and flipping will be performed using a Mezei-type precession coil $\pi$-flipper [16]. Uniaxial polarisation analysis will be achieved using a wide-angle Si-windowed $^3$He spin filter cell, to be contained in a vessel also holding the sample environment. The paper is structured as follows: in section 2, we will give an overview of the history and the aims of the project. Then, we will provide a more detailed description of the simulation and design of the polarised neutron components in section 3. The expected performance will be given in section 4, along with a discussion of the possible scientific applications and the relative merits of three-directional ($xyz$) versus uniaxial polarisation analysis. We will conclude in section 5.

2. Project overview

Longitudinal polarisation analysis, in which only the longitudinal components of the beam polarisation with respect to the external field are measured [1], requires means to polarise, analyse, and manipulate the spin state of the neutron beam; the classic implementation on a triple-axis spectrometer comprises a polariser and a flipper before the sample, followed by a set of coils to rotate the beam polarisation into three orthogonal directions, $x$, $y$, and $z$. Following scattering from the sample, the beam then passes through another flipper and an analyser before being detected. This setup permits measurement of the 12 (longitudinal) cross sections of the type $\sigma_{\pm \pm \pm}$ and $\sigma_{\pm \mp \mp}$, where $\alpha = \{x, y, z\}$. These have the general form:

\[
\begin{align*}
\sigma_{\pm \pm \pm} &= \sigma^{nuc} + \sigma_{\pm | \pm \pm}^{{mag}} \frac{1}{3} \sigma^{si} \pm \sigma^{nm} \\
\sigma_{\pm \mp \mp} &= \sigma_{\pm | \mp \mp}^{{mag}} \frac{2}{3} \sigma^{si} \pm \sigma^{chi}
\end{align*}
\]

where $\sigma^{nuc}$ is the nuclear (and isotope incoherent) cross section, $\sigma^{si}$ the spin incoherent cross section, and $\sigma^{mag}$ the magnetic cross section, which is sensitive to both the direction of the incident polarisation and the scattering vector $Q$; non-spin-flip processes $\sigma_{\pm \pm \pm}$ measure the component of the Fourier-transformed magnetisation parallel to the polarisation and perpendicular to $Q$, while spin-flip processes probe the component perpendicular to both the polarisation and $Q$. Finally, $\sigma^{chi}$ and $\sigma^{nm}$ are terms which arise from non-collinear structures and nuclear-magnetic interference, respectively. Separation of the cross section components from these cross sections is generally possible.
On a multi-detector spectrometer, the second flipper is usually dispensed with as a result of the difficulty of flipping over a wide scattering angle, meaning that the final polarisation analysis system consists of a polariser, a flipper, a guide field, and an analyser. As such, only the six of the twelve cross sections i.e. $\sigma_{+\alpha+\alpha}$, $\sigma_{+\alpha-\alpha}$, etc. can typically be measured. Nonetheless, if certain assumptions are made on the form of the magnetic scattering, separation of the cross section components is still possible using either so-called $xyz$ (three-directional) polarisation analysis [9, 11], or the more recent 10–point technique, devised for instruments which integrate over the detector height [17].

LET was designed with a polarisation analysis option from the outset; the beam was originally polarised by a $^3$He spin filter, with the $^3$He gas polarised in situ via the spin exchange optical pumping (SEOP) method [18]. The spin filter and associated optics were mounted on a trolley on rails to facilitate easy changes between the polarised and unpolarised configurations. The $^3$He gas in the polariser, and hence also the neutron beam, could furthermore be flipped by applying an adiabatic fast passage NMR pulse to the spin filter cell. Analysis of the scattered beam was to be achieved using wide-angle $^3$He analyser cells filled remotely by a metastable exchange optical pumping (MEOP) filling station. However, test experiments with the SEOP polariser showed that its maximum $^3$He polarisation $P_{He} = 65\%$, combined with the removal of approximately 1 m of guide, resulted in large losses in flux, in excess of 90% across the wavelength band of LET. As a result of the limitations of the SEOP polariser, and because the moderator spectrum on LET is colder than originally envisaged, it was decided to replace the original polariser by a supermirror-based device, which exhibits superior performance at all but the shortest wavelengths. This device consists of a two-channel tapering guide with each channel containing a pair of polarising FeSi supermirrors arranged in a “V” formation, and its design is detailed in Section 3.1.

The replacement of the $^3$He polariser also means that flipping has to be performed directly on the neutron beam. The length of the new polariser, as well as the large stray field produced by the permanent magnets used to saturate the supermirrors, severely restricts the alternatives in this respect. Indeed, the default option for a white beam with a large cross-section, the adiabatic radio-frequency (RF) flipper, consumes too much space and is sensitive to field inhomogeneities. As such, we choose a much simpler precession coil (or Mezei) flipper, which can be made less than 10 mm thick and is only sensitive to field gradients of $O(10^{-2}|B|)$. Despite the robustness of the device, the large stray field produced by the polariser still requires careful shielding, and since a precession coil only flips a single wavelength for a given current, the current needs to be ramped in time. Solutions to both of these issues will be detailed in Section 3.2.

The polarised beam is to be transported to the sample by a set of permanent magnet guide fields followed by a set of coils oriented perpendicular to the beam direction; in other words, the magnetic field throughout the instrument will point in the $z$ (up) direction, meaning that non-adiabatic rotation of the neutron polarisation is less of a concern for the present implementation, as compared to cases where full three-direction polarisation analysis is attempted. The coils will also generate a homogenous holding field across the volume of the $^3$He spin filter analyser cell, as demonstrated in Section 3.4.

The $^3$He spin filter analyser presents a significant challenge; previous tests of wide-angle composite Si/pyrex cells for use at 3 atm resulted in a high failure rate. To mitigate the risk of cell failure and to simplify their manufacture, we will reduce the operating pressure to $\sim 1$ atm, and maintain the cells (as well as the sample environment) in 1 atm of inert gas by placing them in a vacuum-tight vessel, to be installed inside the LET sample tank. This leads to cell dimensions which are compatible with both the wavelength band of LET and the homogenous region of the holding field, as shown in Section 3.3. We will aim to reduce the background generated by the cell by using polished Si windows for all parts of the cell which transmit the neutron beam. Any remaining scattering from the cell and the sample environment will be
removed by a radial oscillating collimator.

The layout of the new polarisation analysis option for LET is summarised in Figure 1. The supermirror polariser and precession coil flipper are located ahead of the final set of disc choppers, approximately 3 m before the sample position. In the sample area, the coils will be suspended on a rack which also supports the inner vessel, which is to hold the \( ^3 \)He analyser cell and the sample environment. The collimator will be installed on existing supports in the sample tank.

3. Design studies
In this section, we will provide a more detailed description of the components either under design or construction. Some components, like the radial oscillating collimator, have not yet been designed, and we therefore defer their description to a future paper.

3.1. Polariser
With recent advances in polarising supermirror technology, it has become possible to polarise a broad neutron wavelength band in a relatively short distance. This is crucial for LET, where only 882 mm is available to accommodate both the polariser and the flipper. To determine whether this space is sufficient to provide a reasonable neutron polarisation and transmission, we consider a transmission-based device consisting of a tapered supermirror guide containing two pairs of polarising supermirrors arranged in a “double-V” formation (Figures 1, 2). The walls of the device are linearly tapered by 0.29° horizontally and 0.22° vertically, like the focusing guide section the polariser replaces, with the cross section dimensions at the entrance an exit being 63.95(H)×40.00(W) mm\(^2\) and 57.96(H)×32.15(W) mm\(^2\). Furthermore, it is considered as coated with \( m = 2 \) supermirror on the sides and \( m = 4 \) on the top and bottom walls, as compared to \( m = 2/3 \) (top/sides) and \( m = 4/4 \) in the preceding and following guide sections. The lower \( m \)-number on the side walls of the polariser ensures the absorption of reflected neutrons of the undesired spin state. An \( m \)-number of 5 was assumed for the polarising supermirrors; this both permits a short device, and is readily attainable using FeSi deposited on an Si substrate. Three device lengths – 732, 782 and 832 mm – were simulated to find a compromise between leaving sufficient space for the flipper and polariser performance. Finally, the necessity of dividing the device into two channels was considered by simulating it with and without a vertical \( m = 2 \) double-sided centre wall. The simulations were carried out for the entire LET instrument from the moderator to the sample position using the McStas package.

The criteria for acceptable performance were a transmission \( T \) of at least 35% (70% of neutrons with the correct polarisation state, here \( |↓⟩ \) with respect to the guide field) and a neutron polarisation \( P_n \) of > 95% across the wavelength band \( 3 < \lambda < 9 \ \AA^{-1} \). This could be achieved for a device of length 782 mm, leaving approximately 70 mm for the flipper when vacuum flanges and windows are included. While a longer device yields slightly better polarisation and transmission, we consider the stray field in excess of what can be easily shielded at the flipper position (see Section 3.2). The neutron polarisation \( P_n \) and transmission \( T \) for a polariser length of 782 mm are plotted versus \( \lambda \) in Figure 3, along with the conventional quality factor \( P_n^2T \). Results are shown for both the design with (two-channel) and without (single-channel) the \( m = 2 \) dividing wall. Starting with the polarisation, the two-channel device clearly performs better than the single-channel version, with the latter showing dips in \( P_n(\lambda) \) at approximately 5 Å and 7 Å. The origin of these features are reflections of neutrons with angles of incidence on the back face of the polarising supermirror \( n\phi_v \) where \( n \) is an even integer and \( \phi_v \) is the angle of the polarising supermirrors [19, 20]. These processes are largely eliminated by the dividing wall.

Despite the significantly improved \( P_n(\lambda) \) of the two-channel configuration, \( P_n^2T \) for the two devices is quite similar, owing to the higher transmission of the single-channel case. Nonetheless, as the differences in transmission are small, we prefer the former due to its more homogenous
Figure 2. Horizontal cross section of the supermirror polariser, with the holding field assembly and housing omitted. The device consists of a horizontally and vertically focusing guide with $m = 2$ and $m = 4$ coatings on the sides and top/bottom, respectively, linearly tapered walls, $m = 5$ FeSi supermirrors arranged in a “double-V” formation, and a dividing wall to create two channels. The device is 782 mm long.

Figure 3. The neutron polarisation $P_n$ (upper panel), transmission $T$, and quality factor $P^2 T$ (lower panel) versus wavelength $\lambda$ for designs of the polariser with (W, solid lines) and without (NW, dashed lines) a dividing wall coated in $m = 2$ supermirror. The design with the wall in place gives a smooth $P_n$ for all wavelengths. The performance of a $^3$He polariser with gas polarisation $P_{He} = 75\%$ is shown for comparison.

performance in $\lambda$. To compare the performances of the present design with the previous $^3$He-based polariser, we calculate $P^2 T$ for the latter assuming a very high $^3$He polarisation of 75\%. Even at the maximum $P^2 T$, $^3$He falls well short of the solid state device. Furthermore, this simple comparison disregards the losses due to removing 880 mm of guide, compared to just 100 mm.

Finally, the FeSi mirrors require a relatively large and homogenous magnetic field to bring them close to saturation. This will be produced by columns of Nd$_2$Fe$_{14}$B permanent magnets sandwiched between soft Fe yokes on the top and bottom of the device. These supply a homogenous field of $\sim 400$ G across the mirrors. However, they also lead to a significant stray field, estimated to be $\sim 100$ G at the flipper position. The implications of this will be discussed in the following section.

3.2. Flipper and guide field

As mentioned previously, the length of the polariser precludes flipper designs which occupy more than 80 mm along the beam direction. When the space constraint is combined with the necessity to operate in the large stray field produced by the permanent magnets around the polariser, even fewer possibilities remain. One of the simplest flipper designs, which is additionally both compact and relatively robust towards stray fields, is the precession coil, or Mezei, flipper [16], which uses a non-adiabatic change of field direction and Larmor precession to flip the beam. Despite its advantages, this type of flipper only flips a single neutron wavelength, and would therefore have to be ramped in synchronisation with the ISIS neutron pulse should the option of repetition rate multiplication be maintained. A precession coil flipper also places material in
the beam, which generates both attenuation and small angle scattering. The latter is, however, mitigated by the 2 m of guide between the flipper and the sample position.

In order to test the suitability of a precession coil flipper for flipping a white beam in the vicinity of a strong magnetic field, we installed a prototype device on the LARMOR instrument, also at TS2, ISIS. The beam was polarised by a “V”-cavity polariser, similar to the one intended for LET, then flipped by the first flipper, an adiabatic RF device. The beam dimensions were defined to be $30 \times 30 \text{ mm}^2$ at the flipper position by two sets of slits. The precession coil flipper, consisting of two coils wound, respectively, along the guide field (the compensation coil) and perpendicular to both this direction and the beam axis (the flipping coil), was installed in a soft iron housing adjacent to the analyser. The latter was a supermirror device consisting of a single FeSi mirror magnetised by Nd$_2$Fe$_{14}$B magnets, which result in a magnetic field $|B| = 300 \text{ G}$ at the exit of the polariser. The transmitted beam was measured for all configurations of both RF and precession coil flippers on and off, with the flipper at three positions along the beam axis; $L_{af} = 45 \text{ mm}$, 100 mm, and 200 mm from the analyser entrance. The flipping efficiency of the precession coil and RF flippers for each configuration was then extracted from the formula [21, 22]:

$$f_{PC} = \frac{I^{00} - I^{01} - I^{10} + I^{11}}{2(I^{00} - I^{10})}$$ (2)

$$f_{RF} = \frac{I^{00} - I^{01} - I^{10} + I^{11}}{2(I^{00} - I^{01})}$$ (3)

where $I^{ab}$ are intensities integrated over the entire detector measured for all configurations of RF/precession coil $(a/b)$ flipper on/off $(0/1)$. The RF flipper was found to be close to 100% efficient across the entire wavelength band (not shown). At $L_{af} = 45 \text{ mm}$, where the field at the downstream face of the flipper was 50 G, the beam was found to be nearly depolarised at the detector (not shown). This was ascribed to steep field gradients in the region of the flipper. Moving the flipper away to 100 mm or 200 mm, however, performance was found to be acceptable, with peak $f_{PC}$ values ranging between 94 – 99% for the former and 98 – 100% for the latter.

In order to quantify this more precisely, we turn to the wavelength-dependence of $f_0$. To fit the curves, we recall some simple results, which, although relatively well known, are not often shown in the literature. Firstly, the time-evolution of the neutron wave-function in the direction perpendicular to the beam and the initial neutron polarisation (which is $|\downarrow\rangle$ for a transmission-based polariser) is

![Figure 4. Flipping efficiency $f_{PC}$ versus wavelength $\lambda$ and flipping current $I_f$ for a flipper-analyser distance of $L_{af} = 20 \text{ cm}$. The expected $f_{PC}(\lambda) \propto \cos^2(\lambda)$ is obeyed for all $B_f$. The first ridge of high $f_{PC}$ corresponds to a $\pi$ flip, and the following ridge $3\pi$.](image-url)
Figure 5. Wavelength-dependence of the precession coil flipper efficiency $f_{PC}$. Curves are shown for three values of $I_f$, with fits to equation 6 indicated by solid lines.

\[
\begin{pmatrix}
\psi_1(t) \\
\psi_2(t)
\end{pmatrix} = \exp\left(\frac{-i\omega_L t}{2}\sigma_x\right) \begin{pmatrix} 0 \\ 1 \end{pmatrix}
\]

(4)

\[
\begin{pmatrix}
\psi_1(t) \\
\psi_2(t)
\end{pmatrix} = \begin{pmatrix} i \sin\left(\frac{\omega_L t}{2}\right) \\ \cos\left(\frac{\omega_L t}{2}\right) \end{pmatrix},
\]

(5)

where $\sigma_x$ is the $x$ component of the Pauli matrices and $\omega_L = -\gamma N B_f$ is the Larmor precession frequency of the neutrons in the field $B_f$ generated by the flipper coil. The probability of finding a spin up after passing through the flipper is thus $P(\psi_1) = |i \sin\left(\frac{\omega_L t}{2}\right)|^2 = \sin^2\left(\frac{\omega_L t}{2}\right)$.

Substituting $t_f = \lambda m_N L_f/h$, where $t_f$ is the time spent in the flipper and $L_f$ is its thickness, and including the absolute flipper efficiency $f_0$ (with $(1 - f_0)/2$ defined as the depolarisation due to the flipper), the wavelength-dependence of the efficiency at a particular $B_f$ is expressed as

\[
f_{PC}(\lambda) = \frac{1}{2} - f_0 \left[ \frac{1}{2} - \cos^2\left(\frac{\gamma N m_N B_f L_f \lambda}{2h}\right) \right]
\]

(6)

\[
= \frac{1}{2} - f_0 \left[ \frac{1}{2} - \cos^2\left(CB_f \lambda\right) \right],
\]

(7)

where the constants have their usual meanings, and, assuming the flipping coil approximates a solenoid, $B_f = 4\pi n [\text{cm}^{-1}] I_f [\text{A}] / 10$, where $n$ is the winding density of the coil and $I_f$ is the current. As can be seen from the fits to this expression for $L_{af} = 200$ mm, shown in Figure 5, agreement is excellent at all values of $B_f$. The effect of moving the flipper from $L_{af} = 200$ mm to $L_{af} = 100$ mm is twofold: the efficiencies $f_0$ (Figure 6) show a broad dip down to 94% around $\lambda = 5$ Å, and there is an offset between the expected peak and actual peaks in $\lambda$. This is probably due to components of the analyser stray field along the flipping field adding to that generated by the coils.

The above measurements and analysis show that a precession coil is a viable option as a broad-band flipper, but also that it has to be shielded carefully from stray fields. In addition,
it is clear that to flip a white beam (or discrete wavelengths in the case of repetition rate multiplication mode), the current through the flipper needs to be both synchronised with the ISIS pulse and ramped in time. Taking the derivative of Equation 6 with respect to $\lambda$, the maximum flipping efficiency is given by the condition $\lambda_{\text{max}} = \pi/2CBf$, and the form of the ramp is found to be

$$I_f(t) = \frac{5}{2} \frac{L_{mf}}{\gamma_N n L_f t},$$

where $L_{mf}$ is the distance from the moderator to the flipper. Some preliminary tests using a signal generator amplified by a power supply and triggered by the ISIS pulse have indicated that excellent performance ($f_0 > 97\%$) at all wavelengths is possible (not shown).

Returning to the issue of stray fields, the field at the exit of the LET polariser will exceed that of the analyser in our test experiment, and $L_{af}$ will only be $\sim 7$ cm. To solve this problem, a model of the flipper and its environs was built in Radia [23], which calculates the magnetic field generated by an assembly of magnetic (both current-carrying and permanent) components using analytical approximations. The model comprised the holding field environment for the polariser, a $\mu$-metal screen (relative permeability $\mu = 5 \times 10^4$) with an aperture to reduce the stray field at the flipper position, the flipper itself, several movable permanent magnets to fine tune the magnetic field, and the three existing soft iron guide field housings following the flipper (Figure 7).

The criteria used to optimise the model were threefold: ensuring adiabatic passage of the neutrons by avoiding sharp gradients in the angle $\theta$ (corresponding to the polar angle between the $z$ and $x,y$ components of the magnetic field), minimising $\theta$ in the region immediately before flipping (denoted $\theta_{pf}$), and maximising the field homogeneity in the flipping region to allow for complete guide field $B_g$ cancellation and accurate rotation of the neutron spin. An additional limitation was imposed by the ohmic heating of the compensation coil combined with its proximity to the Nd$_2$Fe$_{14}$B guide magnets. Increasing the winding density of the copper wire to avoid overheating results in a higher neutron flux loss. On the other hand, aluminium wire can be used for its transparency to neutrons, but in turn suffers from lower electrical conductivity and deterioration under high temperatures, limiting the current. Designs where the guide field exceeded a set threshold ($\sim 60$ G) were therefore disregarded.

The best solution was found by a combination of algorithmic optimisation for a simplified model containing only the flipper and polariser, as well as manual movement of the flipper and magnets for the full model. This solution includes four cube-shaped Nd$_2$Fe$_{14}$B magnets (24x24x24 mm$^3$), magnetised along the guide field and placed near the corners of the soft iron housing of the guide field section immediately following the flipper. Combined with position and strength adjustments of the guide magnets further down the beam, a near-complete cancellation of the stray fields was achieved in the simulation. The remaining variation in the $z$-component of the field $B_z$ (around 3 G, as to compared to the total field $|B| = 58$ G) along the $z$-axis in the flipper will be remedied by slight adjustments of the winding density of the compensation coil. To test the validity of our simulations, a partial model of the flipper was constructed (Figure 7, bottom left), and its magnetic field mapped using a Hall probe. The results of these measurements, which are shown in Figure 7, agree almost quantitatively with the corresponding simulation, giving confidence that the final device will perform as expected.

To estimate the setup’s performance, the standard deviation of $B_f$ across the beam cross-section $\delta B_f$ was calculated. As the Larmor frequency scales linearly with $B_f$, the ratio $\delta B_f/B_f$ corresponds to the fractional error in the rotation angle. As shown above, the expectation value of the rotated polarisation state (here $|\uparrow\rangle$) then depends primarily on the cosines squared of the total rotation and the field direction ahead of the flipper.
Figure 7. (top) A side view of the model used to optimise the magnetic field environment around the flipper. The polariser is on the left, separated from the flipper by a $\mu$-metal screen. The guide field section immediately following the flipper contains several permanent magnets to compensate the stray fields at the flipper position (bottom left). The best solution yields a flipping efficiency $> 99\%$. (bottom right) Comparison of measured and simulated component of the magnetic field parallel to the flipping field around the beam centre for a model of the flipper containing only the surrounding guide field section and magnets. The slight discrepancy is due to a small spatial offset of the Hall probe.

$$P(\psi_2) \simeq \cos^2 (\theta_{pf}) \cos^2 \left[ \frac{\delta B_f}{B_f} \right].$$

The values obtained from the simulations are $\theta_{pf} = 0.035$ and $\delta B_f = 0.03$ G, which gives expectation values $P(\psi_2) > 0.998$ across the desired wavelength range. The losses due to non-adiabatic passage are negligible and are therefore disregarded.

3.3. $^3$He spin filter cells

To analyse the scattered beam on LET, we require a device capable of covering the large solid angle subtended by the detector banks. As mentioned in the introduction, wide-angle solid state analysers are associated with considerable complications in both data collection and analysis, and $^3$He spin filters therefore remain the preferred option, despite the fact that these only perform optimally for a single wavelength. The dimensions of the $^3$He spin filter cells are a compromise between the dimensions of the sample environment, the minimum wavelength $\lambda_{min}$ admitted by the polariser (see Section 3.1), and the maximum admissible pressure in the cell. For $\lambda_{min} = 3$ Å, the pressure length $pl$ required to maximise the quality factor $P_n^2 T$, where

$$P_n = \tanh (0.0733p\lambda P_{He}) = \tanh (OP_{He})$$

(10)
Figure 8. Three dimensional model of the envisaged $^3$He spin filter cell. The cell covers $180^\circ$ in the horizontal plane and $\gamma = \pm 30.2^\circ$ in the vertical direction.

Figure 9. A side view of the sample area, including the coils, cell, collimator, and inner tank. The latter will maintain the $^3$He spin filter and the sample environment under 1 atm of inert gas, such as $^4$He or Ar.

and

$$T = \exp (-O) \cosh (OP_{He})$$

(11)

is $\sim 9$ bar cm. The inner radius is determined by the maximum radius of the sample environment, 40 mm for the cryostat we are currently adapting for exclusive use with polarised $^3$He. For a cell with 5 mm walls operating at atmospheric pressure, the outer radius would thus be at least 140 mm (allowing 5 mm clearance between the cell and cryostat tail), and the height at least 200 mm. As will be shown in the following section, it is difficult ensure a homogenous magnetic field (and hence long $^3$He field gradient lifetime $T_{1,\text{grad}}$) across the volume filled by a cell of these dimensions, especially given the spatial constraints the LET sample space imposes on the holding field coils. On the other hand, previous experience with glued cells suggests that these may be operated at up to 1 bar overpressure. Provided the cell can be contained in a vessel at atmospheric pressure, this allows us to reduce the gas length to 6 cm, much more amenable to long cell lifetimes. The latter may be achieved by the use of an inner vacuum vessel (Figure 9), also containing the sample environment, which fits inside the LET sample tank. The vessel will be filled with a gas with small neutron scattering and absorption cross sections (e.g. $^4$He or Ar) to reduce attenuation of the beam. An additional advantage of this arrangement is that the sample tank vacuum does not need to be broken, which should facilitate rapid cell changes.

Regarding the spin filter cell material, single crystal silicon is both compatible with both long lifetimes and a low background at small scattering vector $Q$. provided care is taken with respect to the orientation of the panels. This is a large advantage over cells constructed of quartz or GE180 glass, where the background is broadly distributed in $Q$ [24], and centred around the elastic line, hence potentially impeding studies at low neutron energy transfers. Given that the cell will intercept the direct beam, an absorbing cadmium strip will be fitted on the inner radius of the cell to cover the beam cross section. This will, however, restrict the scattering angle $\theta$ in the horizontal plane to $|\theta| \geq 11^\circ$. A three dimensional model of the proposed $^3$He spin filter analyser cell is shown in Figure 8.

The $^3$He gas is to be polarised in the FLYNN filling station, installed at ISIS in 2011. FLYNN operates using the metastable exchange optical pumping (MEOP) method, whereby
metastable $^3$He atoms generated by a high-voltage discharge are directly polarised by circularly polarised laser light, before transferring their polarisation to ground state atoms via collisions [25]. Following recent improvements, FLYNN is now producing polarised $^3$He gas with an excellent $P_{^3\text{He}} \sim 75\%$.

### 3.4. Holding field

As discussed in the previous section, polarisation analysis on LET will be achieved using wide-angle $^3$He spin filters. Due to the large size of the spin filter cells (60 mm path length, at least 170 mm tall), the magnetic field environment around the cells must be carefully designed to minimise $^3$He polarisation losses from magnetic field gradients. These losses may be quantified by the inverse relaxation time (or lifetime) $1/T_{1,\text{grad}}$

$$\frac{1}{T_{1,\text{grad}}} = \frac{D}{p} \left[ \left( \frac{\nabla B_x}{|B|} \right)^2 + \left( \frac{\nabla B_x}{|B|} \right)^2 \right]$$

(12)

$$\frac{1}{T_{1,\text{grad}}} = \frac{D}{p} \left( \frac{\nabla B_t}{|B|} \right)^2$$

(13)

where $D/p$ is the $^3$He gas diffusion constant, which is $6700/p$ cm$^{-2}$ hr$^{-1}$ at a pressure of $p$ atm [26]. The terms in the brackets represent the square of the transverse field gradient $\nabla B_t = \nabla \sqrt{B_x^2 + B_y^2}$ normalised to the norm of the field $|B|$. Thus, to achieve a 150 hour lifetime for a cell at $p = 1$ atm, the maximum allowable average transverse field gradient over the cell is approximately $1 \times 10^{-3}$ cm$^{-1}$.

The exceptionally large detector coverage on LET presents another issue; designing a coil-set with a horizontal opening of 180° is trivial, but covering $\gamma_{\text{max}} = \pm 30°$ in the vertical direction without shadowing the detector is more difficult. For example, the well-known Barker and Braunbek coil geometries [27] have fixed openings close to $\pm 10°$.

In order to optimise the coil geometry, we again employed the Radia software for Mathematica [23]. Three configurations were simulated; a simple pair of Helmholtz coils, four coils arranged in two symmetric pairs, and six coils arranged in three symmetric pairs. All coil configurations but the Helmholtz configuration were optimised using the internal Mathematica simulated annealing algorithm. The figure of merit to minimise was

$$F = w_B |B_c - B_i| + w_G \left( \frac{\nabla B_i}{|B|} \right)_{\text{max}} + w_A \left( \frac{\gamma_A}{\gamma_{\text{max}}} - 1 \right),$$

(14)
Figure 11. Field and field gradient maps for the optimised four coil configuration. The left panels show the absolute field in the XY (top) and XZ (bottom) planes, while the right panels show the relative gradient of the transverse component of the field. The red lines indicate the position of the cell, with the inner and outer walls denoted by dashed and solid lines, respectively.

where $B_c$ is the field at the centre of the cell, $B_i$ is the desired field, here 10 G, $(\nabla B_i/|B|)_{\text{max}}$ is the maximum relative transverse gradient along either the inner or outer wall of the cell, and $\gamma_A$ is the opening angle of the coils, as defined in Figure 10. The weights $w_B$, $w_G$, and $w_A$ were adjusted for each model to yield an acceptable compromise between the three terms.

The best solution was found for four coils arranged in two symmetric pairs; the main pair with 269 windings at a separation of 426 mm, and the compensation pair with 77 windings and the current reversed at 361 mm (Figure 10). The figure of merit was $F = 2.0$ as compared to 16.4 for the second best model, the six coil configuration, and 27.4 for the third best model, the Helmholtz configuration. This is largely a consequence of the fact that the four-coil configuration allows a full vertical view of the LET detector, i.e. $\gamma > \gamma_{\text{max}}$. In addition, the magnetic field profiles (Figure 11) confirm that the field gradients for this model are excellent, with the entirety of the cell held in a gradient of $< 5 \times 10^{-4}$ cm$^{-1}$. Indeed, the maximum transverse gradient observed is $4.3 \times 10^{-4}$ cm$^{-1}$, corresponding to a $T_{1,\text{grad}}$ in the region of 800 hr. However, adiabatic transport of neutron polarisation from the polariser to the sample position remains to be simulated.

4. Expected performance and applications

The guide system on LET has recently been upgraded with a focusing snout to provide a smaller, more divergent beam at the sample position. The resulting gain in flux is a factor 3 versus the previous version of the instrument; the unpolarised flux at $\lambda = 5$ Å and 2% resolution is now $4.4 \times 10^4$ n cm$^{-2}$ s$^{-1}$, while for a relaxed resolution of 4%, this increases to $8 \times 10^4$ n cm$^{-2}$ s$^{-1}$. The high flux, coupled with the exceptionally low instrumental background, make LET ideally suited for polarisation analysis. Indeed, the high transmission of the polariser will deliver a polarised flux at the sample position of $1.5 \times 10^4$ n cm$^{-2}$ s$^{-1}$ at 5 Å and 2%, similar to the unpolarised pre-upgrade instrument. Additional losses will of course be incurred in the $^3$He analyser. For example, if the gas pressure is matched to an incident wavelength of $\lambda = 7$ Å ($E_i = 1.67$ meV), the peak transmission will be 33% at the elastic line (assuming an unpolarised scattered beam), but only 24% at 0.6$E_i$. As such, in cases where inelastic features are of interest, it may be desirable to adjust the optimum wavelength to that of the scattered rather than the incident beam.
Other cold time-of-flight instruments with current or planned polarisation analysis capabilities include MACS, NEAT, and HYSPEC. Initial experiments on HYSPEC and MACS, both of which currently operate in uniaxial mode, have shown that polarised experiments are viable on a reasonable timescale, typically one week. On these instruments, polarisation and analysis are achieved using two $^3$He spin filter cells and a Heusler reflector and supermirror array, respectively. Contrary to $^3$He, the transmission of a supermirror-based device is typically flat for long neutron wavelengths, perhaps making supermirror-based instruments more suitable for inelastic studies over a broad energy range. Nevertheless, the high combined transmission of the polariser and analyser will make LET perform at a commensurate level with the above instruments once complete.

We finish by debating the relative merits of performing uniaxial and three-directional polarisation analysis on a wide-angle time-of-flight spectrometer. At the outset, we again stress that polarised inelastic neutron scattering is a flux limited technique and that three directional polarisation analysis measurements typically take at least three times longer than uniaxial ones. It is thus hard to envisage completing a three-directional measurement in a reasonable time-frame given current instrument performance. Furthermore, inelastic neutron scattering with three-directional polarisation analysis has typically been applied to single crystals of superconductors (e.g. to separate anisotropic spin fluctuations), conventional and quantum magnets (e.g. to distinguish longitudinal and transverse magnons), and materials with hybrid excitations (e.g. electromagnons). A triple-axis spectrometer is superior to a time-of-flight spectrometer for addressing most these problems, as the excitations probed are typically localised in $(Q, \omega)$ space (quantum magnets with continuum excitations are excepted). On the other hand, time-of-flight spectroscopy with uniaxial polarisation analysis can complement triple axis spectroscopy by surveying large regions of $(Q, \omega)$ space for interesting points to probe in more detail with three-directional polarisation analysis. In addition, for certain systems, uniaxial polarisation provides sufficient information to separate the spatial components of the magnetisation. For example, uniaxial polarisation analysis has been successfully applied to uncover pinch points and other diffuse features in rare earth pyrochlores like Ho$_2$Ti$_2$O$_7$ and Tb$_2$Ti$_2$O$_7$ [28]. We expect future polarised single crystal measurements on LET to be divided between these applications.

For powder samples, it is also debatable how much extra information can be gained by performing full three-directional polarisation. If little or no spin incoherent scattering is present, as in many metal oxide systems, a full separation of the scattering cross section is straightforward from a uniaxial measurement. In some other instances, energy resolution can help separate the cross section components. For example, in a metal-organic magnet, the low-energy dynamics is dominated by quasi-elastic scattering from protons, and there is very little coherent nuclear contribution. The magnetic scattering can then be isolated from the spin incoherent by a linear combination of the spin-flip and non-spin-flip cross sections. Where three-directional polarisation analysis is desirable, as in, for example molecular nano-magnets, the large amount of incoherent scattering from the protons in the organic ligands often makes polarisation analysis considerably less time-efficient than sample deuteration.

Non-magnetic samples, of course, also only require uniaxial polarisation analysis. Recent work on D7 at the Institut Laue-Langevin has identified the promise of polarised quasi-elastic neutron scattering (PQENS), a rarely used technique thus far [29]. Here, polarised neutrons can be used both to separate weak signals from dilute diffusing species in ionic conductors as well as isotopic labelling of functional groups in biological or polymer systems. LET has excellent resolution in energy transfer (down to 8 $\mu$eV at $E_i = 1$ meV) and can cover both several decades in time ($5 \times 10^{-13}$ s – $3 \times 10^{-9}$ s) as well as a broad range in $Q$ using repetition rate multiplication, making it well suited for PQENS. A particularly promising avenue to explore is diffusion of protons, Li$^+$ and Na$^+$ in inorganic battery and fuel cell materials.
5. Conclusion
We have presented design studies for the components comprising the future polarised option on the LET spectrometer, located at the ISIS Neutron and Muon Facility. The polarised neutron beam will be produced by a relatively short “V”-cavity polariser containing $m = 5$ FeSi supermirrors. Flipping will be achieved using a simple precession coil device, selected due to its small size and its robustness to stray field. The wide-angle $^3$He analyser cell, to be constructed of Si, will be maintained in a homogenous magnetic field defined by two pairs of coils wound along the $z$ direction. Fabrication of several of the components has already begun, and completion of the hardware is anticipated in 2017, and the final performance should be similar to instruments at other sources. We have also discussed the relative merits of uniaxial and three directional polarisation analysis, and come to the conclusion that the former will be sufficient for most applications. A major challenge will be the development of software to correct and analyse the large datasets generated by time-of-flight spectrometers.

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