Wide-angle polarization analysis with $^3$He for neutron scattering instrumentation at the JCNS

A Ioffe*, E Babcock, V Pipich, A Radulescu
Jülich Centre for Neutron Science at FRM II, Forschungszentrum Jülich GmbH,
85747 Garching, Germany

E-mail: a.ioffe@fz-juelich.de

Abstract. Polarization analysis is an important technique for polarized neutron scattering as it allows one to obtain the full information about the vector magnetization in the sample that is critically important for detailed understanding of physical properties of molecular magnets, new superconductors, spin electronic and magnetic nanostructures, as well as the self-organization of magnetic nanostructures. In the simplified 1-dimensional version polarization analysis allows for the separation of coherent and incoherent scattering, making it a potentially important technique for studies of non-deuterated biological objects that themselves produce unavoidable background. We compare some of the major considerations between two different methods for the polarization analysis – supermirror based analyzers and polarized $^3$He neutron spin filters and point out when the latter is beneficial from the point of view of our neutron experiments and instrumentation. We will also discuss some specific requirements to such neutron spin filters and summarize the classes of instrumentation where they will be applied at the JCNS. Finally we will describe a successful application for small-angle neutron scattering from a biological sample.

1. Introduction
Detailed understanding of physical properties of new substances such as molecular magnets, new superconductors, spin electronic and magnetic nanostructures, as well as mechanisms involved in their formation, e.g. the self-organization of magnetic nanostructures, requires the full information about the vector magnetization in the sample, i.e. both about the magnitude and the spatial direction of the magnetic field.

Generally speaking, the ultimate task of polarized neutron scattering experiments is to determine the depolarization tensor $D$ that defines the polarization of the scattered beam $P_j$ after the scattering

$$P_j = D P_i$$ (1)

where $P_i$ ($i = x, y, z$) are the values of the three orthogonal components of the neutron spin in the incident beam and $P_j$ ($i, j = x, y, z$) are the values of the polarization measured for $i$-th component of the neutron spin in the incident beam and for $j$-th component of the neutron spin in the scattered beam. The polarization tensor is given by the matrix $[1, 2]$.
To perform 1-dimensional polarization analysis one determines only the polarization $P_{zz}$ (z – the quantization axis) by measurements of spin-up and spin-down scattering intensities. However, even in this simplified form polarized neutron techniques are very useful. For example, 1-dimensional PA allows for the separation of (weak) coherent and incoherent scattering from the sample (see Sect. 4).

Another technique is the so-called full polarization analysis (or XYZ polarization analysis) when one determines the diagonal elements of the matrix $D$ (Eq. 2). In this case 1-dimensional polarization analysis is separately performed for all 3 components (x, y and z, i.e. for two spin directions normal to the quantization axis and one parallel to it, respectively) of the incident beam polarization and the 3 values $P_{xx}$, $P_{yy}$ and $P_{zz}$ are determined. With this method the magnetic scattering cross-sections can be separated from the nuclear scattering in the scattering from the sample in addition to the abovementioned separation of the incoherent and coherent scattering cross sections.

Finally, using the vector polarization analysis (or as it is also called spherical polarization analysis) one can also determine the off-diagonal elements of the matrix $D$ (Eq. 2) and establish the orientation of the magnetic field vector [1, 2]. Moreover, a spread in the orientation of the magnetic field vector leads to a spread in the orientation of the outgoing neutron spin vector causing the depolarization of the incident neutron beam. Thus, the vector analysis of the neutron polarization together with neutron depolarization measurements may provide full information, about the magnetic field distribution (both amplitude and direction) in mesoscopic/nanoscopic magnetic structures.

All these applications require the polarization analysis of the neutron beam scattered by the sample. Dictated by the limited brilliance of neutron sources, a large number of new neutron scattering instruments, both recently built and in the design phase, are equipped with large area position-sensitive detectors to compensate for the low brilliance and/or low scattering intensity by measuring as much of the scattering pattern at once as possible (see [3-6] for some examples). This means that the polarization analysis of the scattered beam should also be performed over the wide scattering angle spanned by such detectors in order to use the instrument effectively.

Moreover, as the new generation of neutron sources (SNS, J-PARC and ESS) are spallation sources, which produce relatively short high intensity bursts of neutrons of a broad energy spectrum, the neutron energy can then be determined via the different source to sample flight times for neutrons of different energies. Thus the neutron instrumentation designed for these sources aims to fully exploit the time structure of their neutron beams and must be able to provide efficient polarization analysis over the full neutron energy spectrum produced in each neutron pulse.

Wide area neutron detectors are used differently depending on the type of measurement performed. The value of the maximum scattering angles $\Omega_h$ and $\Omega_w$ (here indices $h$ and $w$ denote the height and the width) to be covered by the analyzer differs significantly for different neutron scattering instruments (see Table 1). They range from about 3° for high $Q$-resolution small angle neutron scattering (SANS) diffractometers to about 190° for diffractometers and time-of-flight spectrometers. As an example, values of
solid angles \((\Omega_h \times \Omega_w)\) for certain neutron scattering instruments \([7]\) at the JCNS are presented in Table 1.

Besides the rather varied requirements of the solid angles for different instruments, there are also some specific requirements to the required quality of neutron spin analyzers that they should not degrade the general neutron performance of the instrument, in particular they should not spoil the instrument resolution. This leads one to prefer polarized \(^3\)He neutron spin filters \((^3\)He NSF\) over supermirror analyzers in many instances because \(^3\)He NSFs are normally considered not to alter the neutron beam divergence.

**Table 1.** Parameters of the sample-detector parts of different types of neutron scattering instruments operated by JCNS at FRM II \([7]\).

| Detector size (height x width) | Sample-detector distance | Solid angle \((\Omega_h \times \Omega_w)\) |
|-------------------------------|--------------------------|----------------------------------|
| TOF spectrometers \((\text{TOPAS})\) | 2 m x 9 m | 3 m | 45° x 150° |
| Diffractometers \((\text{DNS})\) | 1 m x 5 m | 1.5 m | 33° x 190° |
| Reflectometers \((\text{MARIA})\) | 0.4 m x 0.4 m | 2 m | 11° x 11° |
| Low Q-resolution SANS \((\text{KWS2})\) | 1 m x 1 m | 1 m | 45° x 45° |
| High Q-resolution SANS \((\text{KWS1})\) | 1 m x 1 m | 20 m | 3° x 3° |

However, the neutron optical quality of the \(^3\)He cell material must not be overlooked: in cases where the scattering angle is very small or the detector spatial resolution is very high the scattering from the surfaces of the \(^3\)He NSF also needs to be considered (this will be discussed in Sect. 3).

In this article we will consider different areas where wide-angle polarization using \(^3\)He spin filters is beneficial from our point of view, discuss some aspects of their implementation and give some practical examples. For purposes of the proceedings of this workshop we will give some general conceptual arguments aimed for a broad audience without intending to give a comprehensive coverage of the topic of wide-angle polarization analysis. Further explanations and more detailed discussions can be found in the given references and the references therein. Aiming to discuss polarization analysis for large area position-sensitive detectors, we will not discuss some common applications of polarization analysis (e.g. for three-axis spectrometers) using single neutron detectors that can be accomplished readily using super mirrors or Heusler crystals. Thorough discussions of this topic and comparisons of these methods to \(^3\)He NSFs can be found in \([8, 9]\).

### 2. Neutron polarizing supermirrors vs. \(^3\)He neutron spin filters.

#### 2.1. Polarizing supermirrors

Neutron polarizers/analysers built on polarizing supermirrors (SM) are widely used in neutron scattering instruments, however only for relatively low energy neutrons. The reason for this is that the critical angle of total reflection of neutrons in the direction normal to the surface, \(\theta_c\), for most materials is rather small and is about \(0.1 \lambda^{-1}\ \text{Å}^{-1}\) maximum, where \(\lambda\) is the neutron wavelength. Using interference multilayer coatings made of thin alternating layers of materials such as Ti, Ni, Fe, or Co of different thicknesses, called supermirrors, one can increase \(\theta\) by the factor \(m\). In recent years substantial progress in the manufacturing of polarizing super mirrors has been achieved that allows commercial production of SMs with \(m\)
up to 4.5. An example of the performance of an industrially produced SM with a very high polarizing efficiency of 99 % and a good reflectivity of about 80 % is shown in Fig. 1 [10].

However, in spite of such a high critical angle, still a very large area of SM coating is required for a real multichannel analyzer made of stacks of many mirrors separated by a small space determined by $\theta_c$, mirror length and the beam divergence defined by the incident beam parameters. Moreover, today the flux on the sample for many instruments is increased using focusing guide systems, which further increase the beam divergence. For divergent neutron beams, the angle of incidence on the mirror should be below $\theta_c$ for any neutron in the beam, so that the mirror should be installed at the angle $i=\theta_c - \beta$ ($2\beta$ is the beam divergence).

In addition, SMs suffer from polarizing efficiency losses because of the critical reflection from Si seen on the left hand side of Fig. 1 for $Q_z<0.11 \text{ nm}^{-1}$, where both spin-up and spin-down components are reflected and the beam polarization becomes zero. Practically, the minimal $m$ value that still provides a high polarization of >95 % is $m_{\text{min}}=0.9$, that limits the useful $m$-range to $\Delta m=m_{\text{min}}=3.1$ for the mirror in Fig. 1. Indeed, for the neutron beam wavelength $\lambda=2$ Å the maximal beam divergence (in the direction normal to the SM; the divergence in the plane of the SM can be as large as desired) that corresponds to $m_r$ is $2\beta=\Delta m \lambda \theta_c = 0.62^\circ$ ($\theta_c=0.1^\circ/\text{Å}$) and the incident angle is $i=\beta+\theta_{\text{min}}=0.49^\circ$, where $\theta_{\text{min}}=m_{\text{min}} \lambda \theta_c$. Thus to cover the whole solid angle $\Omega_w$ of the detector one needs $\Omega_w/2\beta$ SMs of the length $L$ required to overlap a neutron beam of width $s$. Since $L=s/\sin(i)$, then $L=117$ cm of SMs with $m=4$ are required to overlap 1 cm of beam width at $\lambda=2$ Å. For a large detector on a TOF spectrometer or diffractometer with $\Omega_w=150^\circ$ one would need about 280 meters of SMs of a height defined by the vertical scattering angle $\Omega_h$ accepted by the detectors and by the sample to analyzer distance. Indeed, if the SM analyzer is midway between the detector and sample for a 2 m detector height, then the total area of SMs amounts to a few hundred square meters. Moreover, as $i\propto \lambda$, this amount is increased inverse proportionally to the neutron wavelength.

While SMs are indispensible for incident beam polarization, where the beam size is limited to the typical size of a neutron guide and the beam divergence is low, and have been successfully used for wide-angle analyzers for cold neutron reflectometers (e.g. NERA [11], HADAS [12], CRISP [13], PRISM [14] etc.) and spectrometers (e.g. D7 [3], DNS [15], etc.),
their use for some of our applications would be difficult. For example, the wide-angle analyzers for the short wavelength neutrons and large area detector of our thermal TOF spectrometer TOPAS, and coverage of large area detectors with sufficient resolution such as our cold neutron SANS, seem to be best served by an approach using \(^3\)He NSFs. For spin flip scattering that is a small fraction of the total scattering, arguments have been made for the use of \(^3\)He polarization analysis on fundamental bases of the obtained data quality achievable with \(^3\)He NSFs, however this is a rather specific discussion that has been covered in prior literature [9] which we will not discuss further here.

2.2. \(^3\)He neutron spin filters

The operation of \(^3\)He NSFs is in a way similar to that of SM neutron polarizers placed in a transmitting geometry: both are filtering out the undesirable spin component, the only difference is that it is absorbed in the first case and reflected out of the beam in the second case. Both of them allow for the undisturbed propagation of the neutron beam – a feature that is tremendously important for high angular resolution applications such as small-angle neutron scattering and reflectometry. Another advantage of the \(^3\)He NSFs is that they allow for the precise neutron beam polarization knowledge (>99.9 %) that is of a great importance for an accurate determination of small spin-flip signals [16, 17].

In contrast to SMs, \(^3\)He NSFs are not subject to the above-described geometrical scaling with the decrease of the neutron wavelength. Their efficiency is determined by the filter opacity that can be controlled by the polarized \(^3\)He gas pressure so that they are rather compact even when used for thermal and hot neutrons (i.e. for \(\lambda<1\ \text{Å}\)). There are some practical considerations such as the safety limits of cells pressurized to over atmospheric pressure and reduction of \(^3\)He \(T_1\) lifetime due to increased dipole-dipole interactions to be discussed below. However, because of their compactness compared to SMs, \(^3\)He NSFs can provide certain practical advantages for the high solid angle coverage at a lower price.

On the other hand, \(^3\)He NSFs are not free from disadvantages. The main drawback is the time decay of the \(^3\)He polarization \(P_{\text{He}}\) in the cell which leads to a lowering of the neutron beam polarization \(P_n\) and transmission over time if the polarized \(^3\)He gas is not refreshed. This decay is caused by some intrinsic reasons such as dipole-dipole interaction of the \(^3\)He atoms, and by the space inhomogenity of the magnetic holding field [18-20]. Further, interactions with the cell’s walls also cause \(^3\)He relaxation in most common instances. The resulting achieved \(^3\)He lifetime is the sum of the three aforementioned sources: interactions with the cell’s wall, field gradients and dipole-dipole interactions can be respectively written as

\[
\frac{1}{T_1} = \frac{1}{T_{\text{cell wall}}} + \frac{7000}{p} \left( \frac{\partial B}{\partial r} \right)^2 + \frac{p}{750}
\]

(3)

Here \(T_1\) is in hours, \(p\) is the \(^3\)He pressure in bar and the quantity squared in the second term is the transverse magnetic field gradient in cm\(^{-1}\). Since the second and third terms of this equation have the opposite dependence on pressure, in the limit where the magnitude of the magnetic term is comparable to the dipole-dipole term the pressure can actually be used to optimize the cell lifetime. 100 hour lifetimes are generally considered to be the lower acceptable limit because at this level one would have to refresh the \(^3\)He polarization on a daily basis in order to retain reasonably good performance in terms of \(P_n\) and neutron transmission [21]. Stray magnetic fields from external magnets reduce performance and, beyond certain limits because of the quadratic dependence of this term, can result in rapid losses of the neutron beam polarization and transmission if care is not taken over the shielding from such sources.
Figure 2. Time-dependence of the $^3$He polarization by in-situ SEOP method [23]. The green/blue lines indicates the $^3$He NSF performance if the same cell were polarized to the same level and then were allowed to undergo normal $T_1$ nuclear spin decay of the polarization with a 200/100 hour time constant respectively.

Indeed, to boost the performance of $^3$He NFSs reduction of the negative factors is required. Clearly one should attempt to reach the longest life-times for the polarized $^3$He gas, so that changes of the neutron beam polarization during the neutron scattering experiment will not be as important and so that the frequency of refreshing the polarization is minimized. However the ultimate solution of the problem is to preserve the $^3$He polarization at a high and constant level. This can be achieved using continuous optical pumping of the $^3$He, i.e. matching the pumping rate with the rate of $^3$He polarization losses such that the rate balance results in a high polarization at constant level [22]. The so called spin-exchange optical pumping method (SEOP) in principle allows one to keep a high polarization for an essentially unlimited time [23]. Recently we achieved a very high on-beam time averaged $^3$He polarization of above 80 % for over a day (Fig. 2), where the time limit was actually imposed by the experiment duration and not the equipment used. In further testing using a larger cell a good $^3$He polarization of 73 % was obtained and maintained for about 80 hours. The measurement which included the use of a sample magnet at up to 1 T and frequent reversal of the direction of $^3$He polarization by the adiabatic fast passage method did not have any effects on the $^3$He polarization or stability. A more detailed discussion about the comparative performance of $^3$He NSFs with constant and time decaying polarization can be found in [24].

We would like to mention that one can overcome in some way the time decay problem although by a regular refilling of the cell with freshly polarized $^3$He gas or by exchanging the cell with a freshly polarized one, thus keeping the averaged polarization over the experiment duration on a reasonable level with the time decaying $^3$He NSF method. This results in a kind of regular (often daily) maintenance of the $^3$He NSF that requires substantial manpower. On-beam polarization in contrast allows a practically maintenance-free operation. Furthermore, neutron experiments performed with $^3$He polarization held at a constant level using the in-situ SEOP method do not need time dependent polarization corrections to analyze their data and will have the highest obtainable time averaged neutron performance for a given set of experimental conditions. However, for certain applications using banana-shaped cells and/or variable magnetic field directions [25-27] it is not apparent how in-situ polarization could be done successfully, thus a local filling approach, or routine change of cells is envisioned.
Figure 3. An example of the SANS scattering curve from elongated cylindrical aggregates (length 2000 Å and radius 50 Å). The curve was measured by parts at different experimental conditions (the sample-detector distance and the wavelength).

3. \(^{3}\)He polarization analyzers for different neutron instruments at JCNS

Now we will consider instrument requests and possible design parameters of polarization analyzers for a few different classes of neutron scattering instruments, namely SANS, reflectometry and wide angle TOF spectrometry.

3.1 Small-angle neutron scattering (SANS)

One can define two limiting cases for the operation of a SANS diffractometer: the highest $Q$-resolution limit and the highest $Q$-range limit (here $Q$ is the momentum transfer). In the first case the aim is to achieve the minimal possible $Q_{\text{min}}$ that allows for studies of large scale structures. As the $Q_{\text{min}}$ of the detector part of the SANS machine is defined by the ratio of the detector pixel size to the sample-detector distance $L$, this requires the latter to be as large as possible. Therefore SANS instruments are usually very long, most commonly about 20 m, or even up to 40 m (D11 at the ILL [28]) that allows for pushing $Q_{\text{min}}$ down to about $10^{-4}$ Å$^{-1}$. However, in this case the $Q$-range available for the measurements that is defined as the ratio of detector size $H_D$ to $L$ (i.e. $H_D/L$) is rather small.

In the second case, the detector is placed very close to the sample, with $L$ down to 1 m, so that the $Q$-resolution of the detector part of the SANS machine is very low, however the $Q$-range is extended to about 1 Å$^{-1}$. In general, the SANS diffraction spectrum is usually taken by three separate measurements covering large $Q$ area with low resolution ($L\to L_{\text{min}}$, $\lambda\to\lambda_{\text{min}}$), than exploring low $Q$-range with high $Q$-resolution ($L\to L_{\text{max}}$, $\lambda\to\lambda_{\text{max}}$) and then one measurement with intermediate resolution ($L=7-8$ m, $\lambda=2\lambda_{\text{min}}$) that overlaps with both previous measurements. In this way one covers up to 3-4 orders in $Q$ with an optimal resolution over the different parts of the SANS curve (Fig. 3).

Now let us consider the problem of polarization analysis using a \(^{3}\)He NSF on such a setup. Principally one can use the same \(^{3}\)He NSF for all 3 measurements, however the requirements to the quality of optical cells are quite different, if not opposite, in the limit...
cases. Imperfections of the cell surface may play a role as a scattering object so that the scattering from the cell material will be a source of background that can overwhelm a small desired signal. Indeed, the cell walls exposed to the neutron beam should have tremendously good surface to minimize the undesired scattering from the surface roughness.

### 3.1.1 Cell material

The most suitable material for $^3$He SEOP cells is GE180 glass produced by General Electric Co [29]. Such cells have been shown to provide the longest lifetime of the polarized $^3$He and are suitable for neutron use because GE180 does not contain boron, which is a strong neutron absorber, like most common glasses do. To check the feasibility of a typical GE180 glass cell for various types of neutron scattering experiments, we carried out a SANS experiment aiming to study the surface scattering from such a cell’s walls. To avoid any artefacts the empty cell blown from GE180 glass, identical to those used for SEOP to polarize $^3$He gas, was placed in the SANS instrument KWS1 [30] of the JCNS that allows for a common vacuum flight path over the whole setup, so that there is no other material, besides the sample, in the neutron beam. Indeed, the scattering curve represents solely the scattering on the cell’s walls without any extra contributions and therefore requires no corrections.

As one can see from the Fig. 4, the cell made of GE180 glass as blown is showing excessive scattering for $Q<0.01$ Å$^{-1}$ and therefore shouldn’t be used in this $Q$-range if the cell is relatively close to the sample and thus far from the detector. However, for $Q>0.01$ Å$^{-1}$ the scattering from the cell is quite small. As the reference level we choose the scattering from 1mm of D$_2$O because it is used in standard buffer solutions for soft matter samples and represents the lowest scattering power of samples to be measured, (noting most samples have a thickness of 2-5 mm depending on concentration). As one can see, the scattering from the cell is even slightly lower than that from 1mm D$_2$O for $Q>0.01$ Å$^{-1}$ therefore any contributions to the scattered intensity in this regime will be small or negligible.

But how about for $Q<0.01$ Å$^{-1}$, especially for the smallest $Q$ of about $10^{-4}$ Å$^{-1}$? After tests of different materials with different surface treatments, we found that Si wafers that underwent a special chemically-mechanically polishing are providing the best result: their roughness is sufficiently small and as one can see from the result of the experiment (Fig. 4) the level of neutron scattering is so low that it may be considered to be negligible within the error bars of this measurement. While Si-windowed cells have been used for off-line polarized $^3$He NSFs when the gas is polarized outside of the cell, to date no cells employing Si windows have been successfully made to employ direct optical pumping of the cell using the SEOP method which requires heating of the cell commonly up to 190°C or higher. Si-windowed cells have been produced for gas polarized outside of the neutron target cell using either MEOP [31], in which the gas is polarized remotely and transferred to the cell through a valve, or with SEOP wherein the Si-windowed cell was connected to a SEOP suitable GE180 cell via a thin polarization transfer tube to allow exchange of $^3$He gas between the two volumes [32]. For the case of the SEOP cell more development is needed before application to neutron scattering. For very high $Q$-resolution, it should not be overlooked that a polarized $^3$He cell could be placed near the detector, thus enabling use of normal GE180 cells. For example, SANS diffractometers KWS3 or KWS2 at the JCNS in the highest $Q$-resolution mode ($Q-10^{-4}$) do not use the standard large area detector, but rather 9 cm in diameter scintillation detectors, which could be covered with one $^3$He NSF cell, of a common diameter for either the SEOP or MEOP method.
3.1.2 Cell size
In Fig. 5 the placement of a $^3$He NSF analyzer in a SANS instrument is shown for two limiting cases. As one can see in the first case of a large $Q$-range (a) the cell diameter should be rather large to span the whole scattered beam. Assuming that cell is installed at distance $l$ from the sample the required cell size $D_{\text{cell}}$ is

$$D_{\text{cell}} = d_{\text{sample}} + H_D l L_1$$

so that $D_{\text{cell}} = 5$ cm for a typical sample size $d_{\text{sample}}=1$ cm, detector size $H_D=1$ m, $l=5$ cm and the sample-detector distance $L_1=1.3$ m. (Here we assumed that the sample is put close to the cell’s wall which is only possible in the case of samples that do not create field gradients at the $^3$He cell position (which is an evident case for soft matter or biological samples). In this case there is also no strict request to the quality of the cell’s walls and therefore GE180 cells as blown can be used without any complications.

For the other case of high $Q$-resolution (b), $L_2$ is about 20 m, so that for the same parameters $H_D$ and $l$, the cell diameter can be much smaller and be practically equal to the sample size, i.e. about 1 cm. However, in this case there is a strong request to the quality of the cell walls’ surfaces that as mentioned could be fulfilled by the use of windows made of chemically polished Si.

Certainly one can try to make a cell that will satisfy both cases, however it seems to us to be most appropriate to use a standard GE180 blown cell with a good lifetime and a small Si windowed cell polarized with a diffusion tube to be discussed below for the second case. A third possible case presented at the end of section 3.1.1 in which for very high $Q$-resolution, low $Q$-range SANS, a cell could be placed close to the detector in which case small amounts of scattering from the cell walls would be unimportant.
3.1.3 Cell polarizing efficiency

Another problem is that the maximal balance of neutron transmission and neutron polarizing power of a $^3$He NSF is achieved when the opacity, i.e. the product of the gas pressure in bar, the cell thickness in cm and neutron wavelength in angstroms is about 30. At opacities much below 30 for commonly obtained $^3$He polarizations, the polarising efficiency of the $^3$He NSF becomes unacceptably low, i.e. $<90\%$. For opacities to far above the optimal opacity, again for commonly obtained $^3$He polarizations, the cell transmission becomes unacceptably low. Indeed, the cell optimized for short wavelength neutrons for the case of the low $Q$ limit (a) (see Fig.5), would be too opaque if used for long wavelength neutrons required in the case (b) resulting in unacceptable intensity losses. In the opposite case, if the cell optimized for the high $Q$-resolution case (b) (i.e. for long wavelength neutrons) is used for the case (a), then the cell opacity for the case (a) (i.e. for short wavelength neutrons) will be too low resulting in unacceptable poor neutron beam polarization. A full discussion of optimal $^3$He NSF opacities is somewhat sample dependent and is discussed in detail in ref. [9].

A possible solution that we are working on is a double-chambered cell made of two different cells connected by a diffusion tube to transfer polarized $^3$He gas between them. One of them will be a large diameter GE180 cell with its thickness and diameter optimized for the case (a), another - a much smaller Si-windowed cell with a smaller thickness and diameter optimized for the low $Q$ limit. The larger GE180 chamber would be heated in an oven and polarized like a normal SEOP cell whereas the smaller Si-window chamber would be outside of the oven, remaining at room temperature, and be passively polarized via. gas exchange with the GE180 cell. In this case, both chambers of the double cell will be also optimized from the point of view of the allowable scattering from their walls'. This idea is in some ways an extension of an idea formed for fundamental physics beam line, and a double cell was created for this purpose, however in this case it was never planed that the GE180 cell would be used for an NSF as well. The cell in that work showed a good $T_1$ relaxation time for the $^3$He implying on should be able to polarize it well, however more testing is needed [33]. Therefore more work needs to be done on the principle of double chambered Si-windowed plus GE180 cells before it can be practically applied.

An example of the possible performance of such device filled with $^3$He polarized at 70 $\%$ is presented in Fig. 6. In this graphic we assumed a rather modest polarization compared to those obtained in normal one-chamber GE180 SEOP cells. Increasing the $^3$He polarization beyond this value would serve to further increase the allowable bandwidth of each portion of such a double chambered cell. As one can see a high dynamic range for the polarizing power over a large lambda range can be realised, which would allow for high-quality measurements for all three experimental conditions shown in Fig.6.
Figure 6. Example assuming \( P_{3\text{He}} = 70\% \) and a double chambered cell with a large diameter GE180 cell optimized for the high Q limit connected by a diffusion tube to a much smaller Si-windowed cell optimized for the low Q limit. In principle a double cell such as this could be effectively used starting at 5 Å and retain good performance at well over 20 Å.

3.2 Reflectometry

In polarized neutron reflectometry (PNR) our scientific cases of interest are mostly studies of magnetic nanostructures, as MARIA will also measure polarized GISANS (Grazing Incidence SANS), and MBE (molecular beam epitaxy) grown layered systems with strong interlayer exchange, both of which require high fields on the order of 1 T or more to saturate the magnetization [34, 35]. Such magnets generate fringe magnetic fields that can result in magnetic field gradients over the cell and in turn lead to a serious reduction in the lifetime of the polarized \(^3\text{He}\). To reduce the influence of such fringe fields, one should try to keep the \(^3\text{He}\) cell far enough away from the magnet and/or design effective shielding of the sample magnet field at the \(^3\text{He}\) NSF position. This however results in a significant increase of the cell size: e.g. the magnetic reflectometer MARIA at the JCNS [36] employs a large position sensitive detector with \( H_{\parallel} = 0.4 \text{ m} \) and \( L_z = 2 \text{ m} \), so that the diameter of the cell should be about 14 cm when placed outside the sample magnet at a distance of \( l = 60 \text{ cm} \) from the sample \( (d_{\text{sample}} = 1 \text{ cm}) \) where the 60 cm represents the minimum distance allowed by our current mu-metal shielded magnetic cavity design [13]. For the scattering measured on a typical GE180 cell we would expect the minimum spot size resolvable due to the scattering from the cell to be \(< 2 \text{ mm} \) at 4.5 Å. As the detector resolution for MARIA is also about 2 mm we do not expect the use of a GE180 cell to appreciably degrade detector resolution. However, for the optical pumping of such a large cell, the hybrid K-Rb SEOP method should be used in order to maximize the efficiency of the lasers used in order to effectively polarize the large 1.5 l volume [37].

3.3 TOF spectrometers and diffractometers

The same considerations as for PNR are valid also for TOF spectrometers and diffractometers requiring ultra wide-angle analyzers as in most studies of magnetic materials or strongly electron-correlated systems, strong magnetic fields are applied. As an example, for the thermal neutron spectrometer TOPAS being constructed by our group, the JCNS at FRM II, a compensated 5 T magnet will be used that has been designed to give a fringe field below
0.1 mT at maximum field at a distance of 60 cm from its center. If the NSF is placed at this position (60 cm), then the volume of a banana-shaped $^3$He cell with a 10 cm thickness and a sufficient height to accept the entire scattered beam would be about 50 litters. To be an effective analyzer for thermal neutrons with $\lambda=1$ Å the $^3$He pressure in such a cell should be about 3 bar, so that about 150 bar-litres of polarized $^3$He will be necessary to fill such a volume.

Though in principle the magnet can either be shielded or further compensated to give acceptable $T_1$ lifetimes, nevertheless the open geometry of such a large magnetic system will hardly provide an ideal magnetic environment for the $^3$He NSF. This will certainly place limits on the maximum attainable lifetimes of the $^3$He polarization. Therefore, a frequent and probably daily refilling of the cell is envisioned. As the required amounts of polarized gas, about 100 bar-litre/day, cannot be produced by any current or planned metastable exchange optical pumping (MEOP) stations to our knowledge, a high output SEOP pumping station is being considered as such a system is currently under development (see e.g. [38]). We do not preclude that eventually higher production rate MEOP systems could become possible as much fundamental work is being done on this area [39, 40]. Consequently any future system cable of polarizing the required volumes would be suitable for this application.

4. Application of polarization analysis in soft matter studies

As was already mentioned in the introduction, using longitudinal or XYZ polarization analysis one can separate different contributions to the neutron scattering cross section [41, 42]. When applied to soft matter studies, the power of both these methods is in the possibility to separate coherent and incoherent contributions. The incoherent scattering in this case is caused mainly by hydrogen atoms which always constitute a significant part of atoms in any polymer or biological molecule with a very large incoherent cross section of 80 barn per atom. The latter should be compared with coherent cross sections of the other most abundant constituents of such molecules: 5.5 barn for carbon, 11 barn for nitrogen and 4.2 barn for oxygen per atom. Therefore, the hydrogen atoms in polymer and biological molecules are strong intrinsic sources of neutron background at the detector. Because the partial incoherent cross section of hydrogen in a molecule is usually much larger than the partial coherent cross sections of the other elements in the same molecule, the intensity of the isotropic incoherent scattering in certain $Q$ regimes is much higher than the intensity of the anisotropic coherent scattering. In other words, the coherent scattering is centered around $Q=0$ and decreasing with a power law $Q$-dependence ranging from $Q^1$ to $Q^4$ (corresponding to the two limits of infinitely long rods and hard spheres respectively) whereas the incoherent scattering is flat such that at high enough $Q$ the incoherent scattering will dominate for any sample. This means that the useful structural information in the diffraction pattern at high $Q$ is covered to various degrees by an overwhelming isotropic incoherent scattering background and the resulting signal-to-noise ratio in the extracted coherent scattering signal is very low.

Moreover, because in this case the incoherent background is caused by the sample itself, the normal differential approach which is to perform two measurements - one with the sample and one without, then subtract these two patterns and to consider their difference as the scattering from the sample is not applicable at all. Straightforward calculations of intensity caused by the incoherent scattering from the sample are not useful because of the multiple incoherent scattering processes that results in the significant, sample geometry dependent, increase of the incoherent cross-section [43].

Additionally, because systematic errors in the absolute calibration of the measured scattered intensity in the units of scattering length (cm$^{-1}$) are not less than 10 %, the
The subtraction of the analytically calculated incoherent background is not possible. However, it is not only difficult to obtain the relevant coherent scattering from the sample, in addition measurements with a strong incoherent background result in an ambiguity of the fit parameters determined from the scattering curve, which we illustrate graphically in Fig. 7. A thorough discussion of the problems in the determinations of protein structures arising from incorrect subtraction of the “baseline” scattering can be found in ref [44]. Here the scattering curve from a protonated structure is presented in a Porod plot (the log(I)-log(Q) presentation). The architecture is analyzed using the empirical Beaucage model [45, 46] that is characterized by three fitting parameters: the Guinier scaling factor \( G \), the radius of gyration \( R_g \) and the Porod exponent \( d \). However, because of the unavoidable incoherent background the fourth fitting parameter, the \( Q \)-independent background intensity \( I_{\text{bkg}} \), must be introduced to describe the scattering pattern over the entire \( Q \)-range covered in the measurement. This results in a strong correlation between this parameter with the power exponent \( d \) that delivers the morphology of the structures. Therefore, changing the assumed level of background \( I_{\text{bkg}} \) one can fit the scattering curve with very different power laws, with \( d \) ranging from -1.6 to -4 (see Fig. 7), that makes an unambiguous determination of the sample morphology impossible. This is especially important in SANS studies of biological structures, where a proper and accurate subtraction of the solvent contribution and the determination of the incoherent background level from the sample becomes particularly critical in the \( Q \)-range above 0.1 Å\(^{-1}\).

A standard solution of this problem is the reduction of the incoherent background by the isotopic substitution of hydrogen atoms with deuterium, which is chemically equivalent but has an incoherent cross section of only 2 barn and a coherent cross sections of 5.6 barn per atom. However, the procedure of isotopic substitution is rather complicated and time consuming, so that not all substances can be prepared in the deuterated form in an amount sufficient for a neutron scattering experiment. Indeed, in some neutron research centers entire laboratories have been created entirely dedicated to the problem of sample deuteration.

![Figure 7](image-url)
As mentioned above neutron polarization analysis is another way to solve this problem. Using a polarized incident neutron beam and analyzing the spin state of scattered neutrons one can distinguish between the coherent and incoherent components [41], effectively raising the signal-to-noise ratio and eliminating the ambiguity from incoherent background. The possibilities of this technique with the aide of a \(^{3}\)He NSF was demonstrated in [48]. Recently we applied it in an experiment carried out on the SANS diffractometer KWS2 of JCNS at FRM II in Garching (Germany). We studied the structure of lactoferrin (a protein with antimicrobial activity) at 1 % protein concentration in a D\(_2\)O buffer solution (pH7). As a non-deuterated protein it is the source of a strong and unavoidable incoherent background.

To provide a wide angle for the polarization analysis the sample was placed close to a polarized \(^{3}\)He NSF allowing it to span a solid angle of about 15 ° corresponding to \(Q<0.35\ \text{Å}^{-1}\). The incident neutron beam was polarized by a transmission super mirror polarizer (the beam polarization was 96.6 % at 4.5 Å). A Mezei-style spin flipper was used before the sample to flip the incident spin with an efficiency of about 97 %. The \(^{3}\)He cell was polarized in the JCNS SEOP laboratory up to the starting polarization of 72 %. The cell was transported to KWS2 and placed in a long shielded, end-compensated solenoid system with a high field homogeneity. A picture of the experimental installation is shown in Fig. 8.

![Figure 8. Solenoid system of the polarized neutron SANS. The sample is placed inside the solenoid system a few cm before the \(^{3}\)He spin filter cell. An overlap of magnetic fields of the polarizer/flipper on the left and the solenoid system allows for the adiabatic rotation of the neutron spin when propagating from the vertical guiding magnetic field into the horizontal magnetic field of the solenoid system.](image)

This \(^{3}\)He NSF provided a high polarization efficiency of 95 % at the beginning of the experiments and remained over 93 % after over 36 hours of measurements. The setup allowed us to obtain a lifetime \(T_1\) of about 430 hours, which is only slightly lower than the lifetime of this cell in ideal conditions, which is 550 hours (see [47] for more details). The level of lifetime performance achieved here would allow us to operate for 4 to 5 days on a single polarization of the cell with a good neutron performance however the data would need to be corrected for the time-decay as stated earlier. Please note that these measurements do not require a flipper after the sample. As the samples have no magnetic scattering all spin-flip scattering events can be interpreted as the result of incoherent scattering processes. A
A detailed description of this method is given in [41, 42].

Errors could occur due to significant changes in the energy of the neutrons transmitted through the sample from inelastic scattering, which will increase the neutron energy, i.e. decrease $\lambda$, the neutron wavelength, of a portion of the transmitted neutron spectrum [49]. Because of the neutron-energy dependent efficiencies of detectors and the $^3$He NSF’s energy dependent polarization analyzing efficiency, a $\tanh(\lambda\Theta P_{He})$ dependence, and transmission, a $\exp(\lambda\Theta)\cosh(\lambda\Theta P_{He})$ dependence, inelastic scattering could lead to errors in the data analysis. Here $\Theta$ is the $^3$He NSF cell opacity or density-length-cross section product, and $P_{He}$ is the $^3$He polarization. For example, in the limit where the polarization analyzing efficiency is about one for all expected $\lambda$, and a “ideal” neutron detector efficiency, the inelastically up-scattered neutrons will be weighted roughly exponentially as a function of their wavelength. However, since the sample is actually a small fraction of the sample plus solvent, and the solvent scattering signal will be measured in both spin parallel and anti-parallel states and subtracted off during data analysis, we should only worry about the effects on the data arising from the inelastic scattering of the sample itself. Thus one should have a reasonable knowledge of the inelastic spectrum from the sample to determine whether or not it will be in the limit where a correction must be made. We will consider these effects carefully in future work. Using time of flight analysis we will study the inelastic spectrums from typical and extreme case samples. Pending the outcomes of this work we will work to develop the required methods and/or corrections for proper data treatment. The data analysis should also account for effects of multiple scattering on the PA, a discussion of which is given in Ref. [48]. A more detailed discussion of the entire SANS data analysis for PA with $^3$He is not a subject of the current article, but may be considered for future work.

A plot of the data obtained is shown in Fig. 9. The black circles are the result of standard measurements with an unpolarized neutron beam (raw data thanks to C. Sill, Forschungszentrum Jülich GmbH). The data are solvent-corrected using a standard approach for the treatment of SANS data from solutions [50]. The red circles are obtained after the data treatment using a polarized incident beam with $^3$He polarization analysis. The

![Figure 9](image-url)

**Figure 9.** SANS patterns from 1% protein lactoferrin in D$_2$O buffer (pH7) [40] solution measured with unpolarized neutrons (black symbol) and polarized neutrons and polarization analysis (red symbols). The red solid line is guided by eye. The $Q$-dependence at high $Q$ can be estimated as $Q^{-3.85}$. 
comparison of these two data sets show the ability to measure the coherent portion of the scattering that is on the order of 100 times below the level of the samples own incoherent scattering, making an unambiguous fit procedure possible. We do not claim that this technique should replace current well-developed techniques to obtain information on the coherent scattering intensity, such as contrast variation methods or to lower the level of incoherent scattering through deuteration, however we feel it will be very useful in difficult cases. The utility of such an approach has been shown in [32] for example.

5. Results and conclusions
Polarization analysis is an imperative technique for polarized neutron scattering when the full information about the vector magnetization in the sample is required. Thus for the detailed understanding of physical properties of molecular magnets, new superconductors, spin electronic and magnetic nanostructures, as well as the self-organization of magnetic nanostructures PA is a must for modern neutron instrumentation. Further it was shown that there are experimental areas of polarized neutron scattering where the polarization analysis using polarized $^3$He spin filters is beneficial, compared to polarizing supermirror analyzers.

In its simplest 1-dimensional version, polarization analysis allows for the separation of coherent and incoherent scattering that could become a useful technique for studies of protonated biological objects whose hydrogen atoms are the source of a strong intrinsic and principally unavoidable background. It was demonstrated in a relevant SANS experiment aimed for the study of the structure of the prorogated protein lactoferrin that the signal to noise ratio can be effectively increased by about two orders of magnitude, thus allowing for the unambiguous interpretation of the experimental data that was impossible without polarization analysis.

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