**Report on first neutron experiments at HMI using a $^3$He-NSF**

To establish the application of $^3$He-NSF’s as polarisers as well as analysers in neutron experiments and to gain experience with this novel technique, investigations making use of them already have been carried out at the triple-axis spectrometer E1 and at the small angle neutron scattering instrument V4, although an in-house supply with polarised $^3$He was not yet available. The particular experiments did not only serve to test the chosen setups, but aimed at scientific impact and thus demonstrated the capability of the apparatus under real conditions.

1. **Investigation on the magnetic and structural disorder of UPt$_2$Si$_2$ at the triple-axis spectrometer E1, using a $^3$He-NSF as a polariser**

From previous investigations UPt$_2$Si$_2$ was known to be a moderately mass enhanced heavy fermion compound which crystallises at 300 K in the CaBe$_2$Ge$_2$-lattice (space group P4/nmm) [1, 2, 3]. Anyhow, the details of the structural distortion had not yet been fully resolved. Therefore a reinvestigation of the structural and magnetic properties of the compound has been performed on annealed crystals in order to determine the effect of crystallographic disorder on the material properties, in particular on the magnetic ground state by means of a detailed neutron scattering study on the AFM phase of UPt$_2$Si$_2$.

![Experimental setup at E1 to investigate the magnetic and structural disorder of UPt$_2$Si$_2$](image)

*Fig. 1: Experimental setup at E1 to investigate the magnetic and structural disorder of UPt$_2$Si$_2$*
The experimental setup is shown in Fig.1. The incoming neutron beam with a wavelength of $\lambda = 2.4$ Å has been polarised by means of a $^3$He-NSF. A cylindrical cell (total length 10 cm, outer diameter 50 mm) made of quartz glass has been used as vessel for the polarised gas. The cell was inside coated with Cs to reduce wall relaxation, resulting in a total $^3$He relaxation time of $\tau \approx 40$ h. The cell was refillable via a glass valve to enable the exchange of relaxed gas by freshly polarised $^3$He. To avoid field-gradient-driven decay of the $^3$He polarisation, the cell was situated inside a transportation unit (see Fig.2), already described elsewhere [4], which creates a magnetic guide field of $B_0 = 8$ G, providing a relative field homogeneity $(\partial B_r / \partial r) / B_0 < 2 \cdot 10^{-4}$ cm$^{-1}$. Thus at the mean pressure of $p = 1.2$ bar which was chosen with regard to the neutron wavelength $\lambda$, the expected gradient-related contribution to the total relaxation rate is $\Gamma_{\text{grad}} < 6 \cdot 10^{-4}$ h$^{-1}$ and is therefore negligible. Due to the axial orientation of the guide field, the outgoing neutrons showed a longitudinal polarisation. Subsequently passing an adiabatic spin rotator, the polarisation was turned up to a vertical direction, effecting an adaptation to the rest of the setup and allowing polarisation analysis by using a Heusler crystal. The neutron spin orientation of the polarised neutron beam could be changed by making use of a spin flipper.

Fig. 2: Transportation unit to house the $^3$He-NSF during both experiment and transportation. At the visible front side, the Mumetal caps which are part of the double-layered shielding are removed to allow an insight into the device. During use they are mounted and will only be detached to take out or to replace the filter cell which is visible on the right.

Concerning the observed relaxation times, the gas filling of the $^3$He-NSF has been exchanged every 12 - 20 hours. Therefore the transportation unit with the filter cell inside
has been removed completely from the setup and was brought to the $^3$He laboratory. Then the cell was taken out and was attached to a rotary vane pump and to a storage cell which was placed in another guide field, respectively. After pumping out the relaxed gas, the filter cell was refilled with polarised gas from the storage cell. Finally the filter cell was detached and was quickly replaced in the transportation unit. Although the field homogeneity during this transfer is less sophisticated, the $^3$He polarisation loss is negligible due to the short duration of the transaction. Containing the filter cell, the transportation unit afterwards was brought back to E1 and was just reinstalled there. A battery-powered supply circuit serves to maintain the field inside the transportation unit also during conveyance.

After each refilling one started with a $^3$He polarisation of $30\% < P_{^3\text{He}} < 50\%$. The $P_{^3\text{He}}$ values have been determined from the flipping ratio of the polarised neutron beam. The concerning neutron polarisation (the values depend also on the actual $^3$He pressure) was $40\% < P_n < 75\%$.

Investigations related to this experiment revealed a crystallographic disorder in the compound in form of strained layers consisting of Pt(2) and Si(2) atoms within the tetragonal CaBe$_2$Ge$_2$ structure. With respect to the magnetic properties, a bulk antiferromagnetic phase transition at $T_N = 32.4$ K has been detected and it has been established that, due to the temperature dependence of the [1 0 0] Bragg peak line width, the magnetic state between $T_N$ and $T_{\text{Ir}}$ consists of magnetic clusters (with a size down to 100 Å) which are embedded in a nonmagnetic matrix, this as result of the crystallographic disorder on the Pt(2) and Si(2) sites. In order to determine if the shape of the magnetic clusters correlates with the type of crystallographic disorder in the compound, we have performed the above-described neutron scattering study on UPt$_2$Si$_2$. The basic idea is to measure the temperature dependence of the line width of a second Bragg peak along an inequivalent direction in reciprocal space (here, the [1 0 2] peak), in order to deduce the correlation length in this direction, using polarised neutrons to suppress the still large structural component of the peak. Presently, data analysis is carried out in order to resolve this issue.

This investigation has been performed in collaboration with S. Süllow from the Technische Universität Braunschweig.

2. Development and test of a local refilling station to make use of a $^3$He-NSF at the SANS instrument V4 and its application in investigations on Co-ferrofluids.

$^3$He-NSF’s are pure transmission filters and thus do not affect the neutron-optical properties of an experimental setup. Hence they are the devices of choice if one intends to analyse the neutron polarisation in cases of large divergence like e.g. in SANS applications. Previous investigations on a Co-ferrofluid at the SANS instrument V4 [5, 6], using a $^3$He-NSF while housing it in the transportation unit mentioned above, proved the technique to work reliably and to be a methodical improvement. Anyhow, this configuration did not fully exploit the
potential of wide-angle polarisation analysis and precluded to make use of the instrument’s standard sample magnet (electromagnet, up to 1 T).

In order to overcome these restrictions, an individual adaptation of the $^3$He-NSF option to the specific requirements of V4 has been effected. Access to a wide detection angle could be achieved by situating the $^3$He-NSF between the pole shoes of the sample magnet, directly behind the sample holder of the instrument (an influence of magnetic samples on the relaxation time constant could be excluded). This configuration provides maximum scattering angles of $\theta_{\text{Max}} \approx \pm 18.6^\circ$ in the vertical direction and $\theta_{\text{Max}} \approx \pm 11.8^\circ$ in the horizontal direction (due to a limitation by the edges of the pole shoes), respectively. In order to achieve a sufficiently homogeneous magnetic field, a new pair of pole shoes has been developed and manufactured with a diameter and a shape sufficient to accommodate both the sample and the $^3$He-NSF inside the highly homogeneous part of the magnetic field. A picture of the rebuilt magnet is shown in Fig. 3.

Fig. 3: Rebuilt electromagnet (sample magnet) of the SANS instrument V4, seen from the downbeam position. Specially shaped pole shoes (black) for a high field homogeneity have been manufactured and mounted. The glass cell in between them is the $^3$He-NSF, the sample holder is placed (in this view) behind the cell. At the right, one can see the upper part of the pneumatically driven lift for a remote removal and replacement of the filter cell.
First of all, the field homogeneity was measured using a Hall sensor and a PC-controlled 3D scanning system, while the $^3$He-NSF was removed from its position. Assuming a perfect cylindrical symmetry, at a typical field strength of 0.6 T, the relative radial gradient was estimated to be $(\partial B_r/\partial r)/B_0 < 7 \cdot 10^{-4}$ cm$^{-1}$. Thus at typical pressures of $p \approx 1$ bar, the expected gradient-related contribution to the total relaxation rate is $\Gamma_{\text{grad}} < 8 \cdot 10^{-3}$ h$^{-1}$.

Fig. 4: Left: diagram of the local refilling station. The filter cell (right, in magenta), placed between the pole shoes of the sample magnet (N, S), is connected via plastic tubing (magenta lines) and a system of valves with a TMP system (in order to remove used gas) and the storage cell, situated in between a pair of Helmholtz coils (red). The valve control circuit controls the gas handling system pneumatically by means of a valve controller. The blue lines represent the tubing for the compressed air. The manually controlled pneumatic lift cylinder serves to remove the filter cell out of the neutron and to replace it there, respectively. Right: photograph of the local refilling station. On the top of the rack in the foreground one can see the Helmholtz coils with the storage cell midway. In the middle of the rack the power supply for the coils is placed with the valve control circuit on its top. At the bottom one finds the TMP unit. The sample chamber with the electromagnet and the $^3$He-NSF inside can be seen just behind the rack.

When applying $^3$He-NSF’s, there is always the need to exchange the $^3$He from time to time for freshly polarised gas, due to the unavoidable relaxation of its polarisation. For the chosen configuration, a replacement of the $^3$He-NSF itself to exchange the $^3$He inside is intricate and difficult. Thus a setup for local refilling (in the following called “local refilling station”) has been constructed which allows to perform this remotely by pumping off the used gas from the
cell and subsequently refilling a new portion of unaltered polarised $^3$He [7]. The layout is shown in Fig. 4.

In the local refilling station, polarised $^3$He is stored in a reservoir, called “storage cell”, which is made of glass GE 180 and has a volume of 1 l. At the beginning of a series of measurements it was loaded up with a gas amount of about 3 bar-l. In order to avoid a rapid depolarisation of the gas, the storage cell is situated in the centre of a pair of Helmholtz coils which serve to keep it in a sufficiently homogeneous magnetic field (0.8 mT). The exchange of $^3$He in the $^3$He-NSF is performed by means of a pneumatically driven gas handling system which is operated via an electronic control circuit. The flow of the fresh gas from the storage cell to the $^3$He-NSF is monitored by using an integrated pressure sensor. The stray fields of the sample magnet and the stray fields of the Helmholtz coils overlap properly to transfer the gas from the reservoir to the analyser cell via small plastic tubes. In case of need, the $^3$He-NSF can be moved away remotely from the neutron beam by means of a pneumatically driven actuator to circumvent an opening of the sample chamber and uplifting the sample magnet during an experiment. In the considered configuration, $^3$He-NSF’s have to be relatively small to fit between the pole shoes of the electromagnet. The filter cells used were made of glass GE 180, too, with a diameter of 50 mm and a length in the beam of 54 mm. They have been kept uncoated inside for the reason of more robustness against maloperation. While during operation the $^3$He-NSF is closed and opened via a pneumatically driven valve, it’s feed pipe also is equipped with a manual glass valve and terminates in a clamp flange to enable a removal from the setup in case of need. In order to avoid magnetic influences on the $^3$He gas which might cause depolarisation, all parts are fully made of non-magnetic material.

It turned out that the high homogeneity of the Helmholtz field as well as the low wall relaxation of the storage cell yield there polarisation relaxation times of about 210 h, allowing to deliver polarised gas over a course of some days of experiments. The electronic control circuit and the valves work properly, and by choosing optimised overflow times and configuring the overlap of the magnetic fields carefully, the polarisation loss during refilling could be reduced to an at least fair value.

The rather small size of the filter cells makes it difficult to achieve a satisfactory relaxation behaviour. The decay of the $^3$He polarisation is strongly depended on the individual cell (what’s not unusual), and therefore a number of cells had been prepared and tested [8]. External NMR tests carried out in low fields (0.8 mT) after cell preparation revealed nonetheless relaxation times $\tau$ up to 40 h, but this value dropped to $\tau = 8 - 9$ h after mounting the cell at the V4 sample magnet, when also applying a comparably low field there. This is much less than one could expect concerning the estimated field homogeneity. A further strong decrease occurred after an increase of the field of the sample magnet ($\tau < 1$ h). It turned out that the main reason for this behaviour was due to the so-called magnetisation of filter cells, confirmed by high-field NMR measurements (1.5 T) at the Universität Mainz. Other possible relaxation sources, like e.g. external magnetic stray fields and external electro-magnetic perturbations at the Larmor frequency, could be excluded. Significant improvements were achieved by proceeding to pneumatically driven glass valves instead of the till-then-used Ti-valves as well as by gluing the clamp flanges to the feed pipe instead of fusing them. Thus relaxation times $\tau = 2 - 3$ h also in the high field range could be achieved.

This still fast decay is not yet been completely understood. Possible explanations could be problems with the field homogeneity caused by ferromagnetic particles (magnetite) in the GE 180 glass, a not perfect cylindrical symmetry of the magnetic field, or impurities in the
used prototype of the $^3$He gas handling system. A detailed discussion of this topic can be found in [9].

A filter cell as described above has been used to carry out investigations on the spin flip scattering intensities of concentrated Co-ferrofluid “MFT” in order to determine the magnetic scattering contribution from it. Therefore a sample of it was placed in the homogeneous field (40 mT) of the electromagnet directly in front of the $^3$He-NSF. In this case, the filter cell chosen featured a relaxation time $\tau \approx (130 \pm 10)$ min. Such rapid decay of the $^3$He polarisation continuously changes the transmissions of the $^3$He-NSF $T^+$ and $T^-$ for nuclear spin polarisations parallel (+) and anti-parallel (-) to the guide field of the electromagnet. The relaxation time is too short to consider the transmissions and thus the analysing efficiency of the filter as being "time-independent" for more than a few minutes.

In general this is accounted to be a disadvantage. In opposite to that common opinion, at HMI a method has been developed which turns the seeming disadvantage into an advantage by exploiting the time dependence of the analysing efficiency to achieve full polarisation analysis without using a second flipper (which would have to be placed between sample and $^3$He-NSF, where anyway is not enough space in the described setup).

With the normal POLARIS setup, two sample intensities

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I^+ = T^+ T^+ + T^- T^- \quad \text{(Flipper OFF)} \quad (1)
\]
\[
I^- = T^+ T^+ + T^- T^- \quad \text{(Flipper ON)} \quad (2)
\]

were measured alternately, into 5-minute files, over a period of 5 hours. Between these measurements, in regular intervals the sample was removed from the beam position by the automatic sample changer, and replaced with an empty slit to measure the transmission of the filter cell, into 1-minute files. Thus two separated but time-linked sets of data were obtained: the sample scattering and the transmission of the filter cell both as a function of measurement time. From the transmission data sub-set, the time dependence of the transmissions $T^+$ (falling from approximately 0.26 to 0.14 within 5 hours) and $T^-$ (rising from approximately 0.08 to 0.12) were approximated with appropriate fit functions. Using these time dependences, then linear regressions of the sample scattering data were performed to extract the spin-flip ($I^+$) and non-spin-flip ($I^-$) components from Eq.(1), and the spin-flip ($I^+$) and non-spin-flip ($I^-$) components from Eq.(2).

![Fig. 5: Raw data (with sample) for flipper "off" (a),(b) and "on" (d),(e) at the beginning (a),(d) and at the end (b),(e) of the measurement time.](image-url)

An example for the scattering data and results of Eq.(2) is given in the figures 5 and 6. Fig.5 shows the change in the raw data during the experiment time due to the filter cell transmission
change caused by the $^3$He polarization decay. Fig.6 shows the spin-flip components $I^+$ and $I^-$ extracted from the raw data set consisting of the starting and ending spectra shown in Fig.1 and the intermediate stages between them. The typical “butterfly” pattern of the flip components caused by the $(\sin \alpha \cdot \cos \alpha)^2$-dependence of the intensity ($\alpha$ is the angle between the scattering vector $\vec{Q}$ and the magnetic field $\vec{B}$) is obvious. A detailed approach to the mentioned investigations and to the novel method described above is given in [10].

![Fig. 6: Scattering contributions $I^+$ (a) and $I^-$ (b) extracted from the experimental data.](image)

The development and construction of the local refilling station, the rebuilding of the sample magnet, the preparation of the filters and their investigation as well as the experiments carried out at V4 have been performed in close collaboration with J. Klenke and W. Heil from the Universität Mainz. The method to exploit the time dependence of the analysing efficiency to achieve full polarisation analysis has been developed in particular by U. Keiderling from the V4 team.

We are grateful to W. Heil and his group at the Universität Mainz for supplying us with polarised $^3$He throughout all experiments described in this report.

References: