PASTIS2 and CROCODILE: XYZ-wide angle polarisation analysis for thermal neutrons

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Abstract. We present a wide-angle device for inelastic neutron scattering with XYZ-polarisation analysis (PASTIS2). PASTIS2 employs a banana-shaped Si-walled 3He-filter for the polarisation analysis and allows pillar-free neutron scattering for horizontal scattering angles 0-100°. The guide field direction at the sample can be chosen vertical or with 45° incremental steps in the horizontal scattering plane. When PASTIS2 is implemented on a polarised neutron beam, the incident neutron spin can be flipped with an easy-to-optimise broad-band adiabatic resonant flipper (CROCODILE) independent of the guide field direction at the sample position. We have tested the performance of this new device on the polarised thermal triple-axis spectrometer IN20 at the Institut Laue-Langevin, equipped with Heusler monochromator and the FlatCone multi-analyser, and discuss its potential for future instruments.

1. Introduction
In spite of intense efforts at all major neutron centers, inelastic thermal neutron scattering with wide-angle polarisation analysis is still outperformed by the point-by-point measurement of triple-axis instruments like IN20. Although wide-angle 3He-spin filter cells have been developed, the achievement of a magnetic field system with sufficient field homogeneity for more than one field direction has been a major challenge. The few wide-angle devices for thermal neutrons that are in operation world-wide (including those with supermirror polarisers) are restricted to a single guide-field direction at the sample, or introduce shadowing pillars in the angular range of the analyser/detector, and/or present such a low polarising efficiency that a triple-axis instrument is probably still more efficient even when it comes to measuring entire sections of the four-dimensional energy-wave vector space. In this paper, we describe a 3He-filter device, PASTIS2, which when combined with the neutron spin flipper CROCODILE presents a major progress towards “parallel” polarisation analysis in the thermal neutron range.

PASTIS2 is the successor of PASTIS1 [1], a first-generation device for wide-angle XYZ-polarization analysis which housed a banana-shaped quartz-glass walled 3He-filter. In PASTIS1, eight vertical pillars intersected the horizontal scattering plane and shadowed a substantial part of the angular range, the quartz glass of the cell introduced considerable background. PASTIS2 provides 135° of free access in the horizontal scattering plane, it allows to choose either a vertical magnetic guide field at the sample or field directions in 45° increments in the horizontal plane, important for polarisation analysis of single crystals [2, 3]. We show that changes in the guide field direction can

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be made with negligible polarisation decay of the $^3$He-filter. The adiabatic fast-passage (AFP) neutron spin flipper CROCODILE permits to flip the incident neutron polarisation independent of the guide field direction at the sample, and flips energy-independent within a broad band of incident neutron energies with an efficiency $f$ above 0.99. We show that it is possible to correct for the finite polarisation and transmission of the $^3$He-filter and their time-dependences in an inelastic experiment, resulting in a final residual “leakage” of only 2.5% of a phonon into a spin-flip channel, which corresponds to an effective flipping ratio of 40. The Cs-coated Si-walls of the $^3$He-filter cell have a transmission of over 0.986, and do not introduce any measurable background. The cell covers the full angular range ($75^\circ$) of the multi-analyser FlatCone. This means that IN20 with FlatCone, PASTIS2, and CROCODILE permits full polarisation analysis over a 75$^\circ$-range of scattering angles simultaneously. While IN20 with FlatCone provides one energy transfer per measurement, PASTIS2 and CROCODILE work for a broad energy band, and could be associated with broad-band incident or final energies as e.g. on time-of-flight spectrometers. There, PASTIS2 and CROCODILE would allow full polarisation analysis for the full range of scattering angles and a broad band of energy transfers simultaneously.

2. Overall Experimental Setup

Equipped with a large horizontally focused Heusler monochromator and analyser, IN20 provides a polarised flux of about $10^8$ n/cm$^2$s (at $k_i$ = 4.5 Å$^{-1}$). IN20 in its single analyser mode allows to measure energy transfers between 1 meV and 100 meV, provides an energy resolution of 0.9 meV and a flipping ratio of typically 27 for elastic scattering at 2.662 Å$^{-1}$. These specifications allow polarised studies of even weak signals like diffuse inelastic spin-1/2 magnetic continua from 50 mm$^3$ single crystals as well as the so-called spherical polarisation analysis in diffraction from e.g. 0.01 mm$^3$ thin-film manganites. For the tests reported here, we used the (polarising) Heusler(111) monochromator, CROCODILE and PASTIS2 (figure 1), and the multi-analyser unit FlatCone [4]. FlatCone covers an angular range of 75$^\circ$ of the scattered neutrons, its 31 vertically bent Si(111) analysers select scattered neutrons with final wave vector $k_f = 3$Å$^{-1}$ independent of their spin state, and scatter them into the vertically displaced detectors. PASTIS2 (figure 2) is a coil device with non-saturated $\mu$-metal cores [5] providing magnetic guide fields in the

![Figure 1](image_url). General layout: Monochromator protection and shutter, CROCODILE, and PASTIS2.
Figure 2. Left: pastis2 with 135° free horizontal access. Right: $^3$He-filter cell, made from Cs-coated single-crystalline Si (cylindric parts) and pyrex (top and bottom).

sample region and houses a banana-shaped Si-walled wide-angle $^3$He-filter. The filter cell fully covers the 75° horizontal scattering angle of FlatCone. pastis2 has 135° free access in the horizontal scattering plane, and allows different field directions at the sample. Throughout this article, we denote with $H||x,y$ horizontal field directions at $-135^\circ$ and $-45^\circ$ to the incident wave vector $k_i$, and with $z$ the vertical field direction. As described below, pastis2 has been modified considerably with respect to its initial version, pastis2.0 [5]. The incident neutron polarisation can now be flipped with the broad-band adiabatic fast-passage (AFP) flipper CROCODILE described in more detail below. We performed experiments on the attenuated direct beam, as well as on a Ge single crystal of about 2 cm$^3$ size, mounted in the $(hh\ell)$-scattering plane inside an Orange cryostat that was kept at 300 K.

3. PASTIS2 design changes

For operation with a horizontal guide field at the sample, the initial pastis2.0 [5] lacked sufficient field homogeneity. The experimentally determined RMS gradient ($>2\cdot10^{-3}\text{cm}^{-1}$) did not meet the expectations from the simulations ($1\cdot10^{-3}\text{cm}^{-1}$) [5], and limited the polarisation decay time $T_{\text{mag}}$ to about 50 hours (h), with an overall $T_1$ of 20 h in situ in early experiments. The original pastis2.0 was improved by replacing the wire-wound x- and y-coils with new coils wound from Cu-tape around the $\mu$-metal cores. The residual field inhomogeneity was measured with a 3D field-mapping robot, and the measured field map was employed in subsequent simulations to design adapted correction coils. A total of 22 correction coils were integrated and optimised via parallel resistances, so that each pastis2 field direction ($x,y,z$) is finally fed by only one power supply. This allows to use standard power supplies to feed pastis2 (otherwise high-stability power supplies would have to be employed). With this optimisation, the relative field homogeneity in the $^3$He-banana cell region was improved by more than a factor 2 for horizontal field directions, from $>2\cdot10^{-3}\text{cm}^{-1}$ to $0.85\cdot10^{-3}\text{cm}^{-1}$. The polarisation decay time due to field inhomogeneities, $T_{\text{mag}}$, measured with a banana-shaped Cs-coated Pyrex glass cell ($T_{\text{wall}}=650\text{ h, 2 bar }^3\text{He}$) in ideal laboratory conditions, nearly quadrupled, from 100 h to 380 h. This translates to an overall $T_1 \approx 100\text{ h}$ in a Si-cell with a typical $T_{\text{wall}}=200\text{ h and 2 bar }^3\text{He}$ as used for experiments. The dedicated $^3$He-filter cells, briefly described in [5], have an inner (outer) radius of 6 cm (12 cm), and a height of 5 cm, figure 2. They provide a vertical take-off $\pm9.5^\circ$. The vertical walls are cut from single-crystalline Si, top and bottom are Pyrex. Pyrex and Si-parts are glued, and the walls are Cs-coated, permitting $T_{\text{wall}}\approx200\text{ h}$. The background
scattering from this type of cell is negligible, in fact, an evacuated cell scatters less neutrons than the corresponding volume of air.

4. CROCODILE - near-adiabatic neutron spin transport and flipping

For horizontal field directions, PASTIS2 presents a sign change of the magnetic field on the incident neutron path that complicates the adiabatic neutron spin transport. Moreover, neutron spin flipping with IN20's standard Mezei-flipper in the incident beam is influenced by stray fields from PASTIS2. The guide field on IN20's monochromator is vertical in order to permit horizontal focusing, and this vertical guide field is not suited for a field flipper. The neutron spin flipping and adiabatic transport difficulties are overcome with the entrance flipper CROCODILE, sketched in figure 3. CROCODILE incorporates a vertical guide field (in prolongation of the vertical guide field of IN20's monochromator protection) that decreases in strength towards the sample position and provides a horizontal guide field at the position where the PASTIS2-field changes sign. The longitudinal gradient $G_0$ of the vertical guide field $B_0(s) = B_0 + G_0s$ along the neutron path $s$ permits the integration of an adiabatic fast-passage (AFP) neutron flipper [6, 7]. In an AFP flipper, a transverse rotating RF magnetic field is applied. The constant RF frequency $\omega_0$ corresponds to the Larmor frequency of the static field in the central region of the field gradient $\omega_0 = |\gamma|B_0(s_c)$, the amplitude $B_1(s)$ is maximum at the resonance position $s_c$, but falls off to near-zero towards the beginning and the end of the static field's gradient region [6]. In a frame that rotates around the static field with $\omega_0$ and travels with the neutron velocity $v$, the neutron spin experiences a field $B_{eff}$ with slowly changing direction,

$$B_{eff}(s) = \left( B_0(s) + \frac{\omega_0}{\gamma} \right) \hat{z} + B_1(s),$$

from $+\hat{z}$ to $-\hat{z}$ for $B_0 > 0, G_0 < 0$, since $B_1 \approx 0$ at the beginning and the end of the gradient region. In the rotating neutron frame the neutron spin follows this effective field adiabatically, and hence in the laboratory system, the spin is flipped after the passage through the flipper.

Apart from the adiabatic condition, the AFP-flipper is independent of the neutron velocity, and hence broad-band. Stray fields will simply shift the "resonance position" slightly, the AFP-flipper is therefore less sensitive to stray fields than a Mezei flipper.

The condition for adiabatic passage,

$$v|G_0| < < |\gamma|B_1^2,$$

is much easier to meet for cold [7] and ultracold neutrons [8] than for thermal neutrons, in particular in presence of geometric constraints for the length of the gradient region. Nevertheless, flipping efficiencies $f > 0.97$ have been reported for thermal neutrons with $\lambda > 0.9 \text{Å}$ with an AFP flipper which had $\gamma B_1^2/vG_0$ of the order of 2 [9]. In CROCODILE the rotating RF-field is replaced by a linearly oscillating one, realised with a solenoid parallel to the neutron beam. This linearly oscillating RF-field can be understood as a superposition of two oppositely rotating RF-fields of half the amplitude, one of which rotates in the sense of the Larmor precession of the neutron spin, the other one has negligible effect on the neutron spin. In CROCODILE, the static magnetic field gradient is about 33 Gm$^{-1}$, the (linear) RF amplitude at the resonance position is 6 G (fig.1), leading to an equivalent rotating amplitude of 3 G. The value of $\gamma B_1^2/vG_0$ ranges between 2.7 and 1.9 (at $k_z = 3 \text{Å}^{-1}$ and 4.3 $\text{Å}^{-1}$, respectively). We demonstrate here, that in spite of the operation in only near-adiabaticity, it is possible to achieve efficient broad-band flipping of thermal neutrons.

5. Data analysis methods

The unpolarised transmission $\tilde{T}$ of the $^3$He-cell, and its analysing power $A$ are given by

$$\tilde{T} = T_0 \exp(-\theta\lambda) \cosh(\theta\lambda P_{3He})$$

(3)
Figure 3. Scheme of CROCODILE. Blue: coils for the static vertical gradient field of the AFP-flipper and adiabatic transport; red: solenoid for the RF-field oscillating with the Larmor frequency; green: solenoid providing a supplementary static guide field parallel to $k_i$.

$$A = \tanh(\theta \lambda P_{3He}),$$  \hspace{1cm} (4)

where $T_0$ is the transmission of the evacuated cell, $P_{3He}$ is the polarization of the $^3$He nuclei, $\lambda = \frac{2\pi}{k_f}$ the wave length of the scattered neutrons, and $\theta$ the $^3$He-cell opacity. At temperature $T = 295$ K, $\theta$ is given by

$$\theta = 0.0728 \frac{\text{Å}^{-1}}{\text{bar cm}} \cdot P_{3He} \cdot \ell,$$  \hspace{1cm} (5)

with the $^3$He-pressure in bar, and the length $\ell$ of the cell in cm. With an incident polarised beam, prepared by a polariser of efficiency $P_i$, a flipper with efficiency $f$, followed by a purely nuclear scatterer with cross section $\sigma^{++}$ (absent or subtracted nuclear spin-incoherent scattering), and analysis with a $^3$He-filter, the non-spin flip and spin-flip intensities are, irrespective of the field direction, given by

$$I_{NSF} = T_0 \sigma^{++} \exp(-\theta \lambda) \left[ \cosh(\theta \lambda P_{3He}) + P_i \sinh(\theta \lambda P_{3He}) \right]$$  \hspace{1cm} (6)

$$I_{SF} = T_0 \sigma^{++} \exp(-\theta \lambda) \left[ \cosh(\theta \lambda P_{3He}) - f P_i \sinh(\theta \lambda P_{3He}) \right].$$  \hspace{1cm} (7)

This holds likewise for measurements with the attenuated direct beam, Ge Bragg peaks, or phonon measurements. All measured elastic intensities were corrected for the non-negligible FlatCone detector deadtime of $\tau = 3.97 \times 10^{-6}$ s.

For the data correction of the inelastic measurements, we need to know $P_i$, $\theta$, $f$ as well as $P_{3He}(0)$ and the decay time $T_1$ for the time-dependent $^3$He-polarisation $P_{3He}(t) = P_{3He}(0)e^{-t/T_1}$. We have determined $P_i$ independently at $k_i = 3$ Å$^{-1}$ on the direct beam, using the “magic box”, a $^3$He-cell with large and known $^3$He-polarisation [10], and an integrated AFP-flipper of efficiency 1 [11]. The value of $\theta$ was calculated from the $^3$He-pressure and cell geometry, eq. (5).
The flipping efficiency of CROCODILE, which includes the polarisation transport in the beam section between shutter and sample, was determined with the CROCODILE as only flipper, and a $^3$He-cell for the polarisation analysis, and is described below. The standard method with a second flipper would have been impractical.

In order to disentangle flipper efficiency and time-dependent polarisation and transmission we measured $I_{NSF}(t)$ and $I_{SF}(t)$ on a Ge(111) Bragg peak, and additionally the intensity with the $^3$He-filter removed, $\sigma^{++}$. The empty-filter transmission $T_0$ of the Si-walled banana-cell was measured separately, $T_0 > 0.986$, and agreed well with the calculated value, $T_0 = 0.992$. The two equations for $I_{NSF}(t)$ and $I_{SF}(t)$ (6,7) contain only two unknowns, $P_{3He}$ and $f$. We used $I_{NSF}$ to determine $\exp(\theta \lambda P_{3He})$, and the measured flipping ratio $R$ to determine $f$

\[
R = \frac{I_{NSF}}{I_{SF}} = \frac{1 + P_1 \tanh(\theta \lambda P_{3He})}{1 - fP_1 \tanh(\theta \lambda P_{3He})} \tag{8}
\]

\[
f = -\frac{1}{R} + \frac{1 - \frac{1}{R}}{P_1 \tanh(\theta \lambda P_{3He})} \tag{9}
\]

A second series of measurements was performed on the attenuated direct beam, with a highly polarised cylindrical $^3$He-filter instead of the Si-banana, for better precision. The beam attenuation was adapted to the low transmission of the $^3$He-cell, the intensity with removed cell was too close to saturating the detector and was not used for the analysis. Since $R$ does not depend on $T_0 \sigma^{++}$, and $f, P_{3He}(0)$, and $T_1$ do not depend on time, it is nevertheless possible to extract $f, P_{3He}(0)$, and $T_1$ from a series of flipping ratio measurements at different times, even without knowing $\sigma^{++}$.

The inelastic measurements were interlaced in regular intervals (about once per hour) by measurements of the Ge(111) Bragg peak, in order to monitor the decay of the polarisation and the transmission.

6. Results

We measured the in-situ polarisation decay in an initial test without CROCODILE, using the attenuated direct beam, a simple solenoid for the adiabatic transport between vertical guide field in the incident beam section and PASTIS2, and a Mezei-flipper, optimized for each field direction and incident energy. Applying the PASTIS2-field horizontally at 135° to $k_i$, $H||x$, we find $T_1 = 51$ h. In a second step we varied the PASTIS2-field direction as fast as possible, rotating between $x$, $y$- and $z$-directions. After 100 rotations we obtain an overall $T_1 = 58$ h, indicating that the rotation itself plays a negligible role for the polarisation decay. Note that employing the CROCODILE which interferes less with the $^3$He-filter cell, we find $T_1 = 70(2)$ h for $H||x$, and $T_1 = 113$ h for $H||z$, to be compared to 20 h with the initial PASTIS2.0.

In order to test the influence of the PASTIS2 stray field on the flipper performance of CROCODILE, we measured the flipping ratio on the direct beam for magnetic field directions every 45° in the horizontal plane as well as for vertical guide field. We employed a $^3$He-cell with high initial polarisation for optimum sensitivity in the polarisation analysis. The raw polarisations spread from 0.8954(15) to 0.8977(15). Corrected for the $^3$He-polarisation decay with time, the polarisations of all directions are identical within 0.4%. This shows that stray fields from PASTIS2 have negligible influence on the flipping efficiency of CROCODILE. For an experiment with full polarisation analysis this means that systematic errors are suppressed to less than one percent of the unpolarised scattering cross section.

Employing the separation method described in the previous section we find the flipper efficiency of CROCODILE is $f_{exp} > 0.999$ for vertical guide field, $f_{exp} = 0.998\pm0.003$ for horizontal guide field direction at 135° to the incident beam at $k_i = 3$ Å$^{-1}$, in very good agreement with the calculated performance of CROCODILE, $f_{calc} > 0.985$ for a beam section of 40x40mm$^2$ and $k_i \in [1.57, 4.5]$Å$^{-1}$. 
Figure 4. Intensity-color maps of the \((hh\ell)\)-plane of Ge at 5 meV energy transfer after correction for the time-dependent polarisation and transmission. The truncated colorscale refers to Counts/5 min, the phonon intensities reach 10000 Counts/5 min. Small residual Ge-Bragg peaks arise from \(2k_i\) and inelastic scattering in the Si-analysers.

By measuring an acoustic phonon emerging from Ge(111), with \(k_i\) ranging from 3.38 Å\(^{-1}\) to 4.0 Å\(^{-1}\) and corresponding incident energies between 23.6 meV and 33.2 meV, we tried to test the broad-band functioning (polarisation transport and flipping) of CROCODILE. For these measurements with finite energy transfer, \(f\) and the incident polarisation were not separated. We corrected the data assuming \(f = 1\) and an incident polarisation \(P_i = 0.9136\) independent of \(k_i\). The resulting effective flipping ratios are about 18-20 for \(k_i\) up to 4.0 Å\(^{-1}\), indicating that CROCODILE indeed works broad-band.

In order to test the feedthrough of a phonon into the spin-flip channel we measured on the Ge single crystal a sector of the \((hh\ell)\)-plane at an energy transfer of 5 meV, with \(k_f = 3\) Å\(^{-1}\). This sector includes the acoustic phonons near (111) and (113). We interlaced the inelastic measurements with flipping ratio measurements on the Bragg peak (111) in order to determine the \(^3\)He-cell polarisation \(P_{3He} = P_{3He}(0) \exp(-t/T_1)\) between start and end-time, \(P_{3He}(0) = 0.776(3)\) and \(T_1 = 70(2)\) h, as described under the data analysis methods. All data were taken with a horizontal guide field \(H||x\) at the sample. The raw flipping ratio decayed from 12 to 8 during the measurements. Figure 4 displays the data after correction for polarisation decay and transmission changes with time. The non-spin flip channel (NSF) displays the acoustic phonons and weaker aluminium powder rings from the cryostat, the spin-flip channel (SF) is essentially empty. The correction assumed \(P_i = 0.9136\) as measured for \(k_i = 3\) Å\(^{-1}\), and the experimentally determined \(f = 0.998\), see above. \(\theta\lambda\) and \(T_0\) play only a role for the overall intensity scale. After corrections, only approximately 2.5% of the phonon intensity leap into the SF-channel, corresponding to an effective flipping ratio of 40. Figure 5 displays the intensities integrated over the entire acoustic phonon ring (integrating over 0.9 Å\(^{-1}\) thick slices in the \((00\ell)\) direction). Here, raw NSF and raw SF label the raw data, while cor. NSF and cor. SF denote the corrected intensities. A dataset taken on the same sample, by simple removal of the cell and changing the monochromator to Si(111), shows that the signal in fully polarised mode is reduced by only an average factor of 12 compared to fully unpolarised mode. This factor contains the difference in monochromator reflectivity as well as the factor 2 that remains even for a perfect polariser/analyser and a hypothetical \(^3\)He-filter transmission of 1.
Figure 5. Acoustic Ge-phonons near (113) (left) and near (111) (right), integrated over 0.8 r.l.u. (0.9Å⁻¹) in the (00ℓ)-direction. Raw NSF and SF denote the raw data, cor. NSF and SF are the corresponding data, corrected for the time-dependent polarisation and transmission.

7. Conclusion
IN20 with FlatCone, the improved PASTIS2, and CROCODILE permits full polarisation analysis for a 75°-range of scattering angles simultaneously, with in situ decay times $T_1$ of the order of 100 h, for variable field directions in the horizontal plane as well as vertical field. We have established a correction method that suppresses polarisation leakage to the level of 2.5%, equivalent to an effective flipping ratio of 40. We have demonstrated that the AFP-flipper CROCODILE works with $f > 0.99$ in a broad range of thermal energies independently of the field direction in PASTIS2, in spite of being operated only near-adiabatically. Due to its insensitivity to stray-fields, such an AFP-flipper, integrated into the monochromator protection, would be extremely useful for polarised experiments in large horizontal or vertical magnetic fields in IN20’s single-detector mode. More generally, both PASTIS2 and CROCODILE are broad-band, and hence could be associated with broad-band incident or final energies as on e.g. time-of-flight spectrometers.

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