Design of Instruments for Polarized Neutrons Using Larmor Labelling Techniques

March 2012

Introduction

Neutron scattering has traditionally been a low-intensity technique, where specialised instruments were built for specialised scientists. With the arrival of high-intensity neutron sources, neutron scattering becomes more accessible to scientists that are specialised in their field of science, rather than in the technique. In this process there is an increasing need for easy and flexible neutron instrumentation. In addition the complexity of scientific problems, which often require switching and tuning measuring ranges "on the spot". A way to address this need for flexibility is to design flexible instrument modules to add on the standard instrument suite of the ESS. With these modules, the instruments will be efficiently adaptable to a broader range of scientific problems.

The development of new experimental methods for neutrons is a field of excellence of the Delft University of Technology. The section Neutron and Positron Methods in Materials (NPM2) and the Reactor Institute Delft (RID) have a long-standing experience in neutron reflectometry, polarised neutrons and Larmor labelling, which led to the development of fundamentally new techniques such as SESANS (Spin Echo Small Angle Neutron Scattering) [1] and novel concepts such as Larmor Diffraction [2]. The Dutch contribution to the design update phase of the ESS will build on this experience and expertise.

In particular the expertise in polarized neutrons is valuable, because the expected performance increase of at least one order of magnitude with respect to existing sources in conjunction with the recent developments in polarizing devices (supermirrors, ^{3}He filters) will make the use of polarized neutrons an attractive option on almost all neutron scattering instruments of the ESS. Beside the traditional use of polarized neutrons in magnetism, polarized neutrons are needed to obtain very high resolution, both spatially and in energy. Also, phase retrieval in reflectometry and separation of coherent and incoherent scattering require polarized neutrons. These "applications" of polarized neutrons should be exploited to a much larger extend at the ESS and will mark a qualitative leap in neutron scattering enabling the full exploitation of the unique capabilities of the neutron spin. High neutron flux in conjunction with polarized neutrons will lead to breakthroughs in science.

This document describes the Dutch contribution to the present design update phase of the European Spallation Source. During the following 2 years phase the design of new concepts for neutron scattering instrumentation will be done. The deliverables will be reports on the feasibility and usability of those new concepts based on detailed simulations using existing Monte Carlo codes [3]. The contribution consists of 4 Work Packages (WP) addressing the potentials of new concepts in a considerable fraction of the traditional neutron scattering instrument suite. The work amounts to 6 man-years: 2 post-docs over 2 years (i.e. 4 man years) and a total of two man-years from the TU Delft staff (W.G. Bouwman, L. van Eijck, C. Pappas, J. Plomp, A.A. van Well).

WP 1: Full Exploitation of Polarized SANS by Spin-Echo Beam Modulation Techniques.

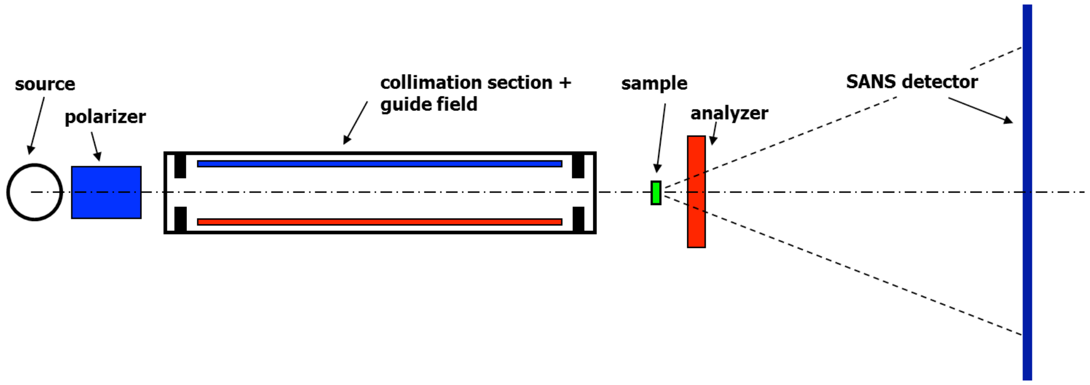

Fig.1: Schematic diagram in side view of the proposed pin-hole SANS instrument with polarized neutrons for which the add-on modules will be designed.

WP 1.1: Polarized SANS

Small angle neutron scattering (SANS) is a powerful technique for the investigation of correlations at length scales from 1 nm up to 100 nm widely used in magnetism, soft matter or for biological studies. SANS instruments may expect large gains – orders of magnitude – at the ESS [4], which will make polarized neutrons an attractive option to all fields of research: in magnetism, where polarisation analysis yields the magnetic structure, but also in all systems containing hydrogen (organic samples and/or the solvent) where polarized neutrons may separate (nuclear) coherent from spin incoherent scattering (see e.g. [5]).

We propose to conceive a dedicated polarised instrument based on the generic "pin-hole" SANS design, with cold neutrons and at a medium distance from the target. The neutron beam will be polarised in the neutron guide, between the target and the instrument, by a super mirror cavity, optimized for a high degree of polarisation and transmission. The polarisation of the scattered beam would be analysed with a polarised ^{3}He filter in combination with a highly efficient RF adiabatic flipper (magic box configuration) to be used as a ≺-flipper for broadband 1D polarisation analysis. The setup illustrated by Fig. 1 will have the space to accommodate spin echo modulation add-ons.

Deliverables

Report on the conceptual design of the instrument on the basis of simulations, including the optimization of the beam delivery system and of the polarizing guide.

WP 1.2: Ultra SANS by adding on Spin-Echo Modulation

Complex matter, soft condensed matter, biology and chemistry are often governed by a hierarchy of length scales, which can stretch from the molecular up to the macroscopic lengths. However, most scattering techniques probe only a limited range of length scales. In order to follow and understand these complex and hierarchical processes it is important to conceive instruments, which cover the largest possible length scales, i.e. from the nanometres to several micrometres in a single measurement. It is a great advantage to reach all these length scales in one instrument, in particular when it comes to fragile biological samples. For this purpose we propose to combine the standard 20 m polarized SANS setup, with a variant of the high resolution Spin Echo SANS (SESANS), which has been developed in Delft [1, 6]. This combination would yield an instrument covering 5 orders of magnitude in attainable length-scales [7]. This unprecedented broad range in combination with polarised neutrons will allow detailed studies of long-range structure and inter-particle interactions in colloidal dispersions, foams, lamellar fragments, supramolecular conjugates, complex mixtures, composites, ‘templates’ and phase segregated systems. These have potential applications in areas such as photonics, drug delivery, gene therapy, catalysis, separation science, and next-generation materials. It will also be possible to determine in the bulk average particle size and interactions on micron scales in opaque samples, a field of great interest for food processing industry. Dairy products are indeed increasingly ‘engineered’, and cooking, freezing, enzyme degradation and the processing of starches all have physico-structural consequences for foodstuffs. Our everyday food is in fact a series of very complex meso-scale systems. Moreover, it will provide important new insights into the porosity of rocks, relevant to pollutant diffusion and to (oil) reservoir engineering, as well as into the transport of pollutants in the aquatic environment by allowing a better understanding of the natural particles and agglomerates in soils, effluents and river sediments to which pollutants sorb.

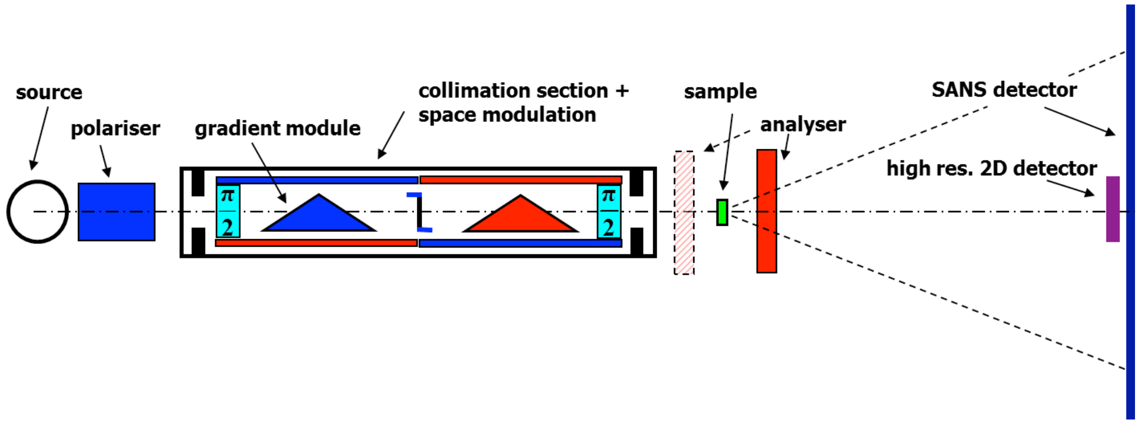

Fig.2: Schematic diagram in side view of the proposed pin-hole SANS instrument in combination with modulated SESANS. The π/2-flippers mark the start and end of the precessions and between the precession elements there is a field stepper. A high-resolution position sensitive detector with low efficiency is used instead of beam stop. This could be stationary and be used in all configurations.

Realisation

Fig. 2 shows a schematic drawing of the Larmor precession device added on the incoming beam of the polarized SANS instrument. This device will create a very fine spatial modulation, which will be measured with a high-resolution position sensitive detector placed instead of the beam-stop at the direct beam. Any SANS by the sample will decrease the amplitude of this modulation [7]. The method is similar to conventional SESANS, but with the great advantage of having all components before the sample, which allows the combination and simultaneous measurements with SANS. To be able to realise this with the most effective design we need to investigate possibilities with magnetized foils and triangular coils [8] used as Larmor labeling and flipping devices. Unfortunately the modulation technique will not practically work when using RF based flipper (time focusing issues).

The magnetized foil based flippers for neutrons are the key component of the monochromatic SESANS in Delft. The advantages of these foil flippers are the well-defined large gradients in precession that can be achieved and the well-defined interface of the different precession regions. This makes them ideal for SESANS applications. However, these flippers function only for one wavelength and for this reason this technology must be modified before it is used at a spallation source, where the wavelength varies with time. There are three options:

1. Use a monochromatic setup, such that the wavelengths for which there are a ≺-flip, 3 ≺-flip, 5 ≺-flip, a (2n+1) ≺-flip can be used as suggested in [9].
2. Generate a periodically moving domain wall in the foil, such that as a function of arriving wavelength always a ≺-flip is obtained [10]. The feasibility of this concept will be assessed on the basis of experiments at the Reactor Institute Delft.
3. Oscillate the angle between the normal of the foil and the neutron beam, so that always a ≺-flip is obtained [11]. Further experiments complemented by McStas calculations will assess the requirements for an accurate movement of the coil.

In this respect the wavelength dependency is not an issue for the triangular coils. In experiments performed in June 2011 [7] revealed the potential of triangular coils [8] composed of current sheets [12] for TOF-SESANS experiments. Development is needed to investigate what the upper field limit will be of such a coil in order to determine its final resolution in this modulation method.

Deliverables

We will make a conceptual design of the combined SANS-SESANS instrument including, the requirements on the magnetic fields [13], the collimator region, Larmor labelling devices and on the detectors. There will be a conclusion on what type of Larmor labelling method is the best option for the ESS on the bases of calculations and limited tests. On the basis of simulations we will deliver a report on the science case for such a setup by comparing the performance of the SANS-SESANS combination with that of dedicated instruments. In addition we will benchmark the proposed spin-echo modulation with other high-resolution options that are presently being developed for SANS.
Monte Carlo simulations will also determine the optimal measurement strategy for realistic samples. This is a non-trivial question to be answered, as the optimal thicknesses and the counting times might be different for SANS and SESANS.

This proposed work package partly overlaps with the German proposed work description I.4. The science case is similar to the Swiss-Danish proposal for a hybrid instrument work package 4.

WP 1.3: Dynamical SANS studies by adding on MIEZE

Modulated intensity small angle neutron scattering (MISANS) [14] combines the features of a traditional small angle neutron scattering instrument with the quasielastic resolution capability of a special version of the spin echo technique—the so-called modulated intensity by zero effort (MIEZE) [15].

MISANS will explore the nanosecond motions of larger entities, which are not only found in biomolecules, polymers, and supramolecular assemblies of nanometer dimensions, but also in metastable states of matter (glasses) or fluids under extreme confinement. In the latter case, much remains to be learnt about how dynamical properties such as viscosity or diffusivity are modified by the confining substrate, as well as how this new paradigm in materials science will be exploited in the design of novel composite materials. The development of a unified conceptual framework for thermodynamic metastability in soft matter transcends purely academic interest, with implications in biological and enzymatic activity or the role of water in living organisms and geological processes and this is a task that is ideally suited for this instrument. Furthermore the study of motions of atoms and ions is of high importance in (nanostructured) ionic conductors, energy storage materials or generally stated, materials for renewable energy applications.

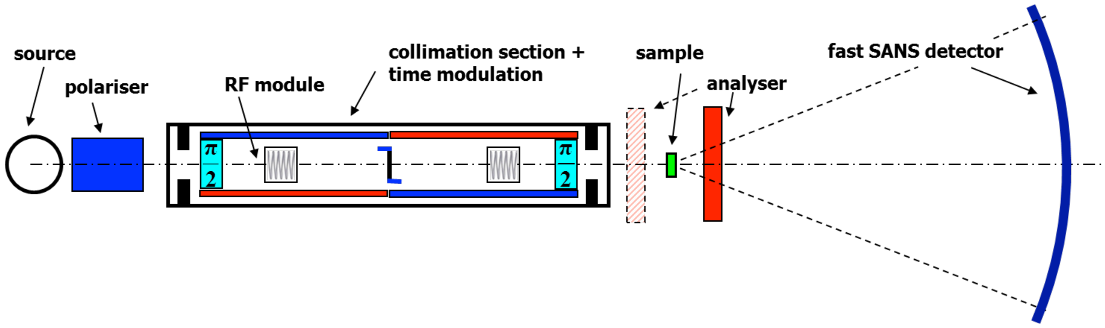

Fig.3: Schematic diagram in side view of the proposed pin-hole SANS instrument in combination with MIEZE. The two RF modules are tuned in such a manner that the intensity at the SANS detector has a fast modulation in time.

Realisation

As illustrated by fig. 3, two RF-modules operated at different frequencies generate a fast modulation at a position sensitive SANS detector. In contrast to most MIEZE realizations, which up to now have used the so-called bootstrap coils and have material in the beam producing parasitic scattering, we propose to explore the use of electromagnets with the RF coils parallel to the beam. Such RF coils with no material in the beam have been developed by the TU Delft and used for the first time at the OFFSPEC reflectometer at ISIS TS2 [16].

Deliverables

Deliverables are a full conceptual design of the instrument, including the requirements on the magnetic fields, requirements on the detectors, definition of the best polarisation devices, precession devices in the collimator range, a conceptual design of the add-ons and their insertion into the collimator, McStas calculations on the performance of the instrument.

A report will be delivered on the science case and performance of the MISANS option discussing the pros and cons of building the instrument with the spin-echo modulation as an add on or as a dedicated instrument.

The report will also benchmark the realisation of MIEZE with RF coils with respect to bootstrap coils and also specify the requirements for the detector.

WP 2: Optimising the Benefits of Spin-Echo Labelling in Polarized Reflectometry

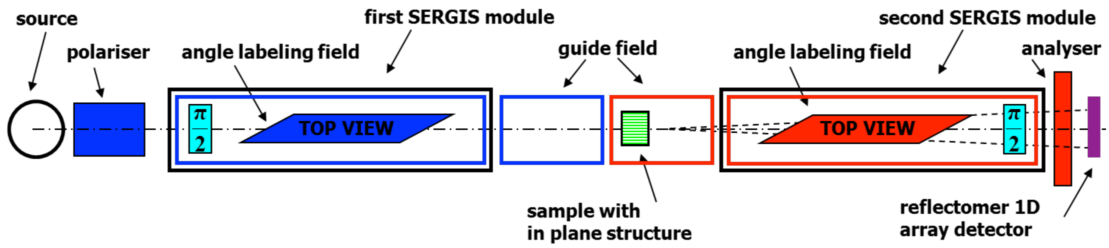

Fig.4: Schematic diagram in top view of the proposed spin-echo option for refectometry.

Specular neutron reflectometry is used for the determination scattering-length density profiles perpendicular to the interface. In-plane structures, in the direction of the neutron beam, will result in off-specular reflection, which can be measured using a position sensitive detector. Whereas the length scale probed by specular reflectivity is determined by wavelength/glancing angle (λ/θ), ranging from a fraction of a nm to ~ 100 nm, the length scale probed by off-specular reflection is determined by λ/θ^2 is of the order several to tens of microns. By using spin-echo angle labelling (SERGIS), in-plane structures, perpendicular to the neutron beam, can be determined in the range from 50 nm – 10 μm [17]. Beside this application, the technique can also be used to separate off-specular scattering from off-specular reflection thus enhancing the resolution of the perpendicular profile of wavy samples [18].

Realisation

A reflectometer with spin echo add-ons based on resonant flippers has been realized at OFFSPEC, ISIS [19]. The long-pulse nature of ESS may lead to additional optimization of the resonant spin flippers. Furthermore, we will investigate whether a specialized instrument has to be designed or the spin-echo components can be constructed in such a way that they can be add-on modules on a conventional reflectometer. Another lesson learned from OFFSPEC is that the magnetic labeling regions, made by RF flippers combined with inclined pole shoes of DC magnets, could gain a factor of three in range if the pole shoe angle was designed for larger inclination angles. There are ideas to improve this for the ESS but this needs more simulations and small scale prototype tests.

Deliverables

Deliverables are a full conceptual design of the instrument, including the requirements on the magnetic fields, definition of the best polarisation devices, precession devices (including ≺/2 flippers, broad-wavelength resonant ≺-flippers), field steppers, a conceptual design of the add-ons, McStas calculations on the performance of the instrument.
A report will be delivered on the science case and performance of the spin-echo options in reflectomery discussing the pros and cons of building the instrument with the spin-echo modulation as an add on or as a dedicated instrument.

WP 3: Exploring Larmor Labelling in Neutron IMAGING

Neutron imaging and neutron tomography are powerful tools for the determination of structures in real space. Those techniques are sensitive to density inhomogeneities, and visualise directly the internal structure of artefacts, such as internal fractures, precipitates, voids or inclusions. In this field, the cold neutrons of the ESS should yield in most cases to a better phase contrast and a much higher resolution.
Neutron imaging and neutron tomography have evolved rapidly in the last years. The implementation of several innovative techniques such as phase contrast [20], dark field [21] or polarized neutrons [22] have significantly increased contrast, resolution and broadened the field of applications. A characteristic of these new techniques is that they are not solely based on absorption, but also use the information from ultra-small-angle neutron scattering and refraction. Along these lines we propose to explore the capabilities offered by Larmor labelling when the gratings used in phase contrast or dark field imaging are replaced by Larmor devices and magnetic fields.

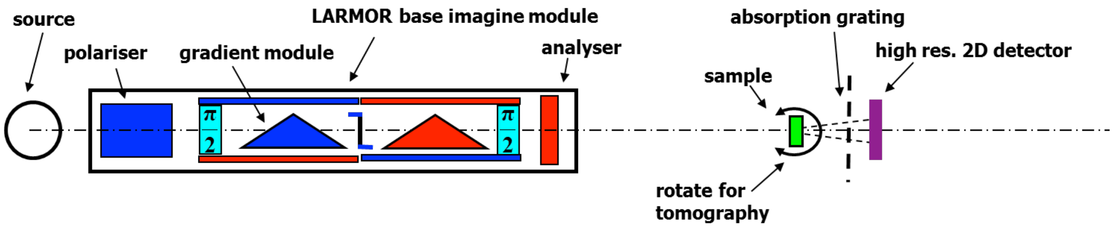

Fig. 5. Schematic view of an imaging station with a Larmor labelling add-on at the incoming beam. Two Larmor precession fields with a gradient create a modulation of the intensity at the absorption grating before the detector. Effectively, they replace the two gratings used in conventional phase contrast or dark field imaging. The sample is on a rotation stage for the tomographic reconstruction.

Realisation

Conventional dark field imaging consists of three absorption gratings. The first two create a line pattern at the position of the third [20-22]. Any small-angle scattering or refraction will decrease the amplitude of the interference pattern with the third grating and will visualise the inhomogeneities in the sample. We propose to create a spatial modulation of the beam using a Spin Echo setup, as shown in fig. 5, instead of the first two gratings. In this setup the periodicity of the modulation will be tuned by adjusting the magnetic fields. This will lead to a much more flexible instrument, where the resolution will be easily adapted to the needs of the experiment, thus optimising the use of neutrons. There is an overlap in Larmor labelling methods with work package 1.2, however, this is a different type of instrument which will have other requirements. It is highly possible that different conclusions will follow for the optimal Larmor Labelling devices compared to WP 1.2.

Deliverables

We will deliver a report on the science case for this instrument and on its conceptual design on the basis of simulations. This will include a choice for the optimal Larmor labelling component. The report will also benchmark the performance of this setup against the "conventional" realisation with absorption gratings.

WP 4: Larmor Labelling in Diffraction: applications of TOFLAR

In diffraction the measured pattern consists of Bragg peaks, as a result of the crystal structures of the phases present. The background between these Bragg peaks often includes diffuse scattered intensity due to inelastic scattering but also to deviations from long-range order, to amorphous, liquid-like or nano-structures. In these studies it is therefore important to distinguish between static and elastic amorphous-like scattering contributions by filtering out the inelastic part. This filtering may be done using a Larmor precession and we propose to investigate the potential of the new technique TOFLAR in diffraction. A successful realization of this new concept will be beneficial to structure investigations of complex construction and functional materials the performance of which depends on the microscopic structure, both crystalline and amorphous. Also alloys, magneto-caloric materials, battery, solar-cell hydrogen storage or fuel cell materials will benefit from it. These functional materials may have a varying amorphous fraction, due to fabrication process parameters, such as annealing or ball milling, or aging during their use, which may be a result of frequent cycling.

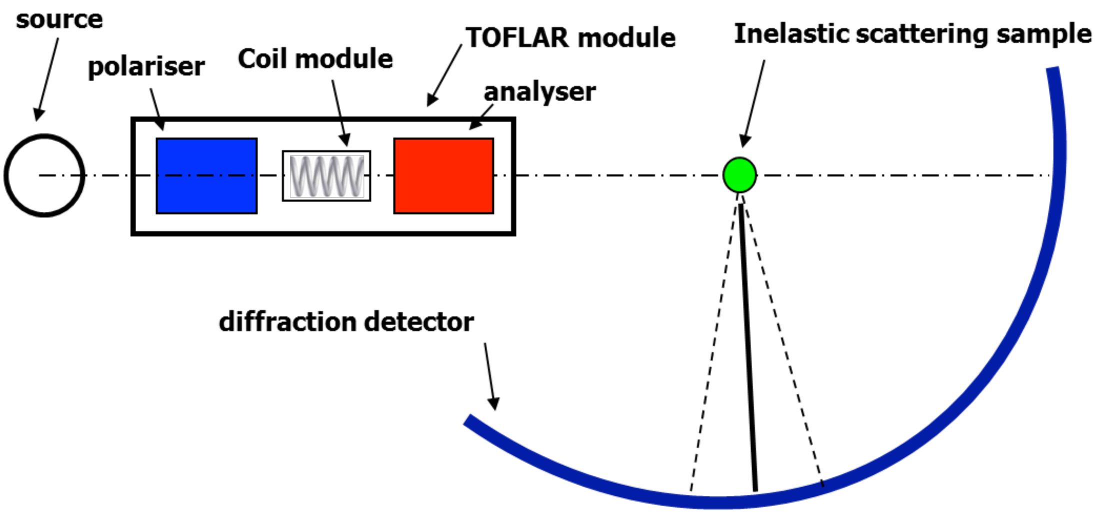

Fig. 6: Schematic drawing (not in scale) of a TOFLAR module introduced at the incoming beam of a powder diffractometer. The module consists of a precession field between a polariser creates an intensity modulation as a function of wavelength and magnetic field. Due to inelastic scattering the amplitude of this modulation will decrease.

Realisation

A Larmor precession device will be positioned at the incoming beam of a high resolution diffractometer, as shown schematically on Fig. 6, and will give rise to a modulation of the detected neutron intensity as a function of the magnetic field (created by the coil module) for each time-of-flight (TOF) channel. The frequency of the modulation will be determined by the corresponding neutron wavelength, the amplitude and damping by the TOF resolution of the host diffractometer. Any inelastic scattering from the sample, which will be outside the TOF resolution will lead to a larger damping. This effect can be followed as a function of applied magnetic field and can be used to separate the elastic from the inelastic contributions. In addition we will investigate the potential of also using TOFLAR in a way similar to a Fourier chopper for high resolution diffraction. The principle of TOFLAR is described in [23] and first experimental results are given in [24].

Deliverables

We will deliver a report on the science case, instrument design and performance for the combination of TOFLAR with diffraction. The report will evaluate the performance, possibilities and limitations of having TOFLAR implemented on an ESS-based high-resolution powder diffractometer. The possibility of using a TOFLAR module as a Fourier chopper [25] will also be evaluated. The calculations as a function of the various instrument parameters will be performed both analytically and by means of Monte-Carlo (McStas [3]) simulations.

References

1. T. Rekveldt et al. Review of Scientific Instruments 76, (2005) 033901
2. M. T. Rekveldt1, T. Keller and R. Golub, Europhys. Lett., 54 (2001) 342
3. http://www.mcstas.org/
4. http://ess-scandinavia.eu/documents/Chapt_5_Vol3upd.pdf
5. E. Babcock et al. , Physica B: Condensed Matter 406 (2011) 2448
6. W. G. Bouwman et al., Physica B-Condensed Matter 350 (2004) 140
7. W. G. Bouwman et al., Physica B-Condensed Matter 406 (2011) 2357
8. R. Pynn *et al.*, Physica B-Condensed Matter 404 (2009) 2582
9. W. H. Kraan *et al.*, Physica B-Condensed Matter 335 (2003) 247
10. M. T. Rekveldt *et al.*, Physica B-Condensed Matter 335 (2003) 164
11. C. P. Duif *et al.*, Physica B-Condensed Matter 406 (2011) 2467
12. M. T. Rekveldt, W. H. Kraan, and J. Plomp, Nucl. Instr. & Meth. A 550 (2005) 54
13. O. Uca, W. G. Bouwman, and M. T. Rekveldt, Nuclear Instruments & Methods A 496 (2003) 437
14. M. Bleuel, K. Littrell, R. Gähler, J. Lal, Physica B 356 (2005) 213
15. R. Gaehler, R. Golub and T. Keller, Physica B 180 & 181 (1992) 899
16. J. Plomp, Spin-echo development for a time-of-flight neutron reflectometer, Ph.D. Thesis, Delft, 2009, ISBN:978-90-9024161-6
17. G.P. Felcher, S.G.E. te Velthuis, J. Major, H. Dosch, C. Anderson, K. Habicht, T. Keller, SPIE 4782 (2002) 164; M.T. Rekveldt, J.Appl.Crystall. 36 (2003) 1301
18. J. Plomp, V.O. de Haan, R.M. Dalgliesh, S. Langridge, A.A. van Well, Thin Solid Films 515 (2007) 5732
19. R.M. Dalgliesh, J.Plomp, V.O. de Haan, S. Langridge, A.A. van Well, Physica B 406 (2011) 2346
20. F. Pfeiffer et al., Phys. Rev. Lett. 96 (2006) 215505
21. M. Strobl et al., Phys. Rev. Lett. 101 (2008) 123902
22. N. Kardjilov et al., Nat. Phys. 4 (2008) 399
23. F.M. Mulder et al, Physica B 276-278 (2000) 132
24. M. Bleuel and A.A. van Well, Physica B 406 (2011) 2478
25. G Badurek and H Rauch in “Neutron Inelastic Scattering 1977”, Vol.1, p.211 IAEA , Vienna 1978