Final report
on the Polarized Techniques Joint Research Activities in the NMI3

Introduction.

The polarized neutrons are particles with the preferential direction of their spin (magnetic moment). They can be considered as a kind of elementary magnet arrows that change their direction when interacting with a magnetic dipolar field in the sample. Their relative orientation can be measured very precisely using neutron polarimeters.
On the other hand, the precession of the around the magnetic field direction allows attachment a “Larmor clock”, which rotation speed depends only on the magnitude of this magnetic field, to every neutron. Such a Larmor labelling opens the possibility for the development of “unusual” neutron scattering techniques, where the resolution does not require the initial and final states to be well selected. This decoupling results in an extremely high energy (or momentum) resolution that is not achievable in conventional neutron spectroscopy (or diffraction) because of intolerable intensity losses.

Wide-angle neutron polarimetry.

The realization of large solid angle neutron polarimetry will allow for the use of large solid angle detectors, which enable the simultaneous data acquisition over a wide range of the transferred momentum. This results in an enormous gain in the efficiency of neutron polarimetric experiments and opens new horizons for detailed understanding of the mechanisms involved in multiferroic compounds, photo induced and molecular magnets, magnetic nanostructures, spin electronic and new superconductors, which are at the forefront of condensed matter research.

A detailed analysis of different possibilities for the wide-angle analyser at the diffractometer has been performed Super 6T2 at LLB. It is found that existing solutions that employ the radial array of polarizing super mirror (SM) analysers are competitive with similar devices based on the 3He spin filters only for long wavelengths, λ>2.5Å, that limits considerably the application of SM analysers in single crystal diffraction and clearly favours the polarized ^{3}He technique for to the single crystal diffraction applications. This technique is based upon the strong spin dependent neutron absorption of the highly polarized ^{3}He gas that makes it an effective neutron spin filter: it is transparent for neutrons with one spin direction and is opaque for neutrons with the opposite spin direction.

FZJ developed a very compact 70cm diameter magnetic system (Fig.1) capable of producing uniform field in three orthogonal directions over a very large solid angle. A pioneer method for manufacturing of doughnut like containers for ^{3}He gas from the dedicated GE180 glass (Fig. 2) has been also developed at FZJ.

Further developments of Larmor labelling methods for inelastic neutron spectroscopy, SANS and reflectometry.

Neutrons may be used to study dynamics on an molecular scale by measuring the neutron’s energy gain or loss during the scattering process from the sample under investigation. Examples of these studies are determining Li diffusion through a battery system or hydrogen transport through a fuel cell. In order to determine the energy change of the neutron, the energy before and after scattering must be known. Present state-of-the instruments select one particular energy of the incoming neutrons, throwing away more than 95 % of the available neutrons, and determine the scattered-neutron's energy by the time-of-flight (TOF) technique.

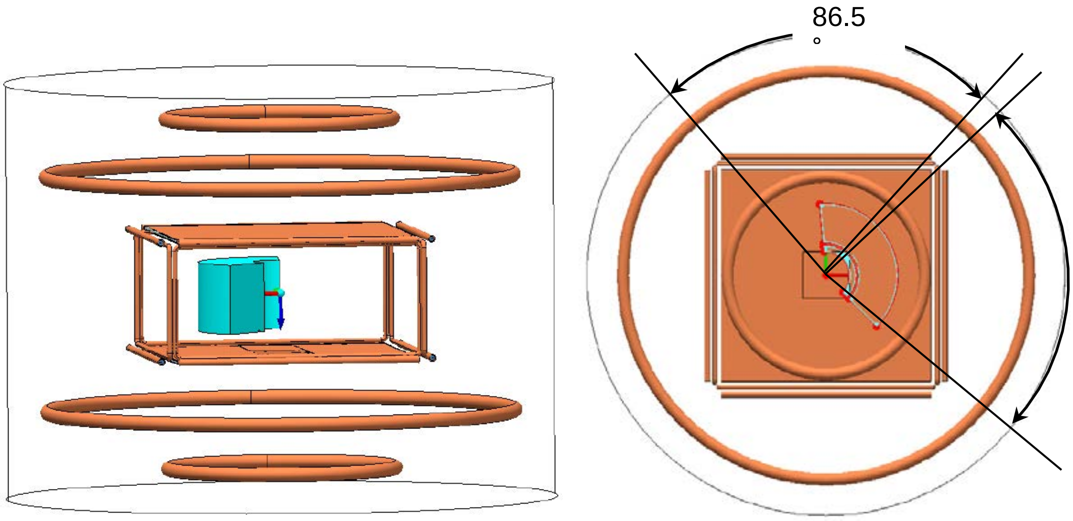


Fig. 1. A very compact 70cm diameter magnetic system capable of producing uniform field in three orthogonal directions over a very large solid angle. The system is calculated to provide unrestricted access to scatter neutron beams over areas of 40 degrees high by 86.5 degrees horizontal, with only 3 degrees of dead area in the horizontal plane between clear areas. This device has been fully modelled using Infolytica "Magnet" finite element software.

Fig.2. The first in the world doughnut shaped cell: outer diameter about 20cm.

In the proposed new TOFLAR technique (TU Delft) the *complete neutron spectrum* will be used and the neutron's energy is determined by a modulation technique: a polarised neutron beam pass through a magnetic field so that the Larmor precession of the polarisation vector, thus labelling the neutron's energy. Combining the Larmor labelling and the TOF technique will result in a quasi-elastic neutron-scattering instrument with high neutron intensity and large accessible range of length and time scales on the molecular level (Fig.3). During the

NMI3 project, the theoretical background is described, a proof-of-principle experiment is performed on a prototype instrument at the Reactor Institute Delft, and computer simulation have been performed.

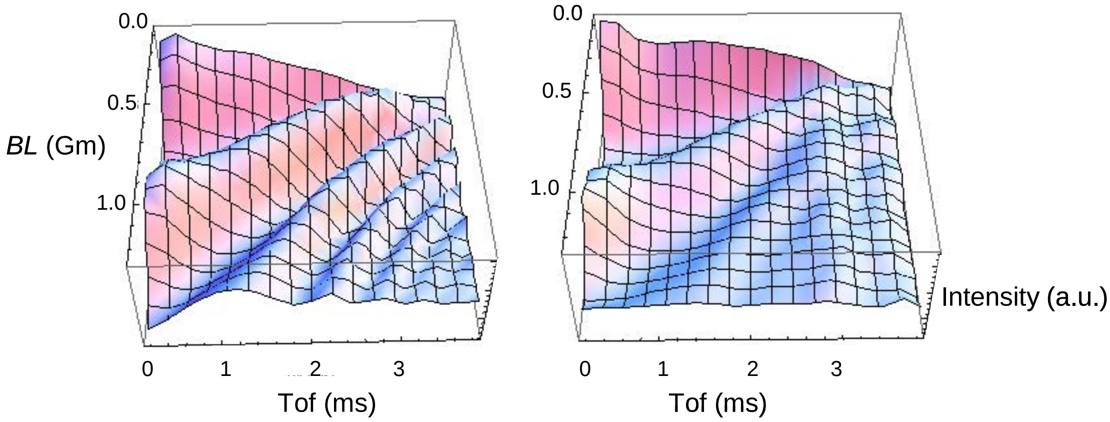


Fig. 3. Measured intensity as a function of time of flight and magnetic field in the Larmor modulator for the straight beam (left) and the scattered beam from a 2-mm-thick acetone sample at a 90^o scattering angle (right).

Neutron spin-echo spectroscopy is a method to analyze slow motions and relaxations on the length scale of atoms, molecules and molecular aggregates. In particular in soft-matter (polymers, proteins, complex fluids like microemulsions, etc.) the combination of neutron diffraction (SANS) and spectroscopy with the contrast generation by selective replacement of H-atoms by deuterium enables deep insights in the structure and mobility of theses systems. The neutron spin-echo spectrometer has a unique resolution to detect very small velocity changes of the scattered neutrons. This resolution is about 3 orders of magnitude higher than that of any other neutron spectroscopic method. Currently the limiting factor to extend the resolution even further is the difference of the factors determining this timing between various possible paths the neutron may take from sample to detector. The variety of paths is necessary to have sufficient intensity, the difference in time-keeping result from differences in the precession magnetic field along the paths. This field drives the rotation of the neutron spins and the differences can be reduced by correction elements in the beam path. The nature and quality of these elements determine the residual errors. To further improve the resolution and the transmission of neutron-spin echo spectrometers new correction elements that have a good transparency for neutrons and at the same time can hold the large currents that are necessary to perform the correction have been developed in FZJ. The challenge is to combine good neutron transmission with high current density and high accuracy of the electric current distribution that is responsible for the corrective action. Both arms (before and after the sample) of the spectrometer contain 3 of these elements, i.e. a complete set consists of 6 elements with different active area. The only practically suited material is aluminium, which combines good neutron transmission with high electrical conductivity. The best theoretical performance is expected from radial current distributions. These were tried first with painstaking effort, however, their practical performance stayed behind the expectations. Thus an idea already employed at the ILL for elements with smaller area was adopted and extended to larger diameter and current density. These so called “Pythargoras coils” consist of crossed current wedges (Fig. 4). Their combined action resembles that of a radial distribution. Despite the fact that their theoretical performance is somewhat non-ideal their realization meets the theoretical expectation and they are the best performing correction elements that currently could be obtained. Following this new design complete sets of coils for the spin-echo spectrometer J-NSE in Garching and the SNS-NSE at the JCNS outstation in Oak Ridge have been produced and are now used there.

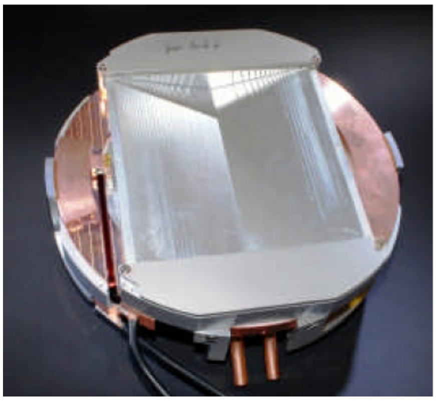
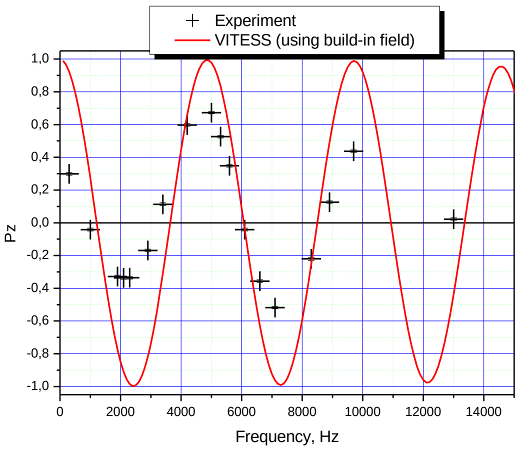


Fig. 4. Crossed Pythargoras coils with thickness modulation and straight cuts each with an active area of 100x100mm.

Monte Carlo simulations are playing a very important role in the context of the design and optimization of neutron scattering instrumentation. An accurate, carefully benchmarked computational model of a neutron instrument allows to underpin the design of new instruments as well as to enhance the efficiency of the existing ones. This makes the development of powerful Monte Carlo instrument simulation codes very important for the progress of neutron scattering research. A significant contribution to these developments has been made in the frame of this project: FZJ in collaboration with Joint Institute for Nuclear Research in Dubna, Russia has extended the possibilities of the simulation software package VITESS allowing for the simulation of spin dynamics in time-dependent magnetic fields. This paved the way to simulations of practically all existing spin-handling devices and Larmor labelling based neutron scattering instruments. An important step was to simulate the performance of real rather than ideal spin handling devices, i.e. those with magnetic field distributions deviating from the assumed ideal ones. Illustration to this is given in Fig. 5, which demonstrates remarkable possibilities of this new software on the example of thin neutron spin turners that are employed for the rotating magnetic fields NSE technique.



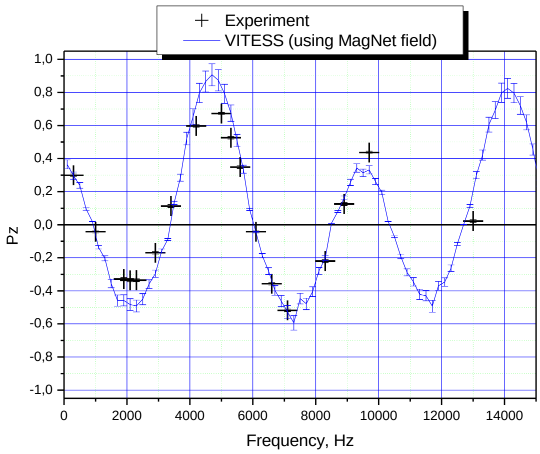


Fig. 5. Frequency dependence of the polarization in comparison with VITESS simulation using built-in (a) sinusoidal and (b) calculated by MagNet magnetic fields.

Development of Large Solid Angle Resonance Spin-Echo

This technique yielding to design and construct Large Solid Angle (LSA) resonance coils for the implementation of a multi detector system on a NSE spectrometer and gain two orders in solid angle detection.
The aim of the project was to design, develop and construct Large Solid Angle (LSA) coils for neutron resonance spin echo spectrometer. In the first option, one can use two flat LSA coils for the primary spectrometer, each of them include two radiofrequency coils inserted in static coils (Fig. 6). The main problems to overcome were to obtain an appropriate

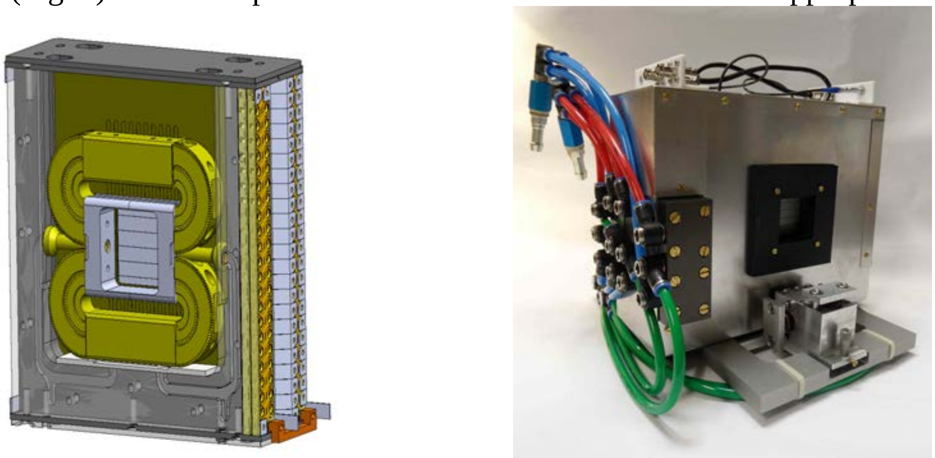


Fig. 6: Flat NRSE coils for the primary spectrometer, left design and right delivered coils. Notice on the designs plans that the RF coils are inserted in the static ones. At maximum field a thermal power of 2 kW must be eliminated.

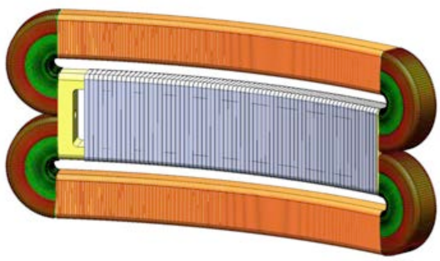


Fig. 7: Curved RF coils for the primary spectrometer.

homogeneity of the field distribution inside the coils, to reduce as far as possible the stray fields outside the coils, to develop an appropriate cooling system (water and air) to avoid mechanical deformation (2kW per coil) and to get an appropriate inductance of the RF coils manage high powers in the frequency range of measurements. For the secondary spectrometer, in order to achieve a measurement over a very wide angle, a curved coil has been developed with the aim to cover simultaneously 15° of scattering angle (Fig. 7). The characteristics of the coils should be similar to the one of the flat coils for the first arm with an additional difficulty related to the fact that we loose a plan of symmetry, and the surface to be covered is wider. In another approach developed in TUM the curved coils are not winded to a body, but cut from one Al piece by means of electrical discharge machining (see Fig.8).

The coil design and the construction were performed at the LLB and TUM and the test has been carried on the spin echo spectrometer G1bis located close to Orphée reactor of the LLB Saclay (Fig. 9).

Fig 8. The first coil cut in Aluminum by electrical discharge machining.

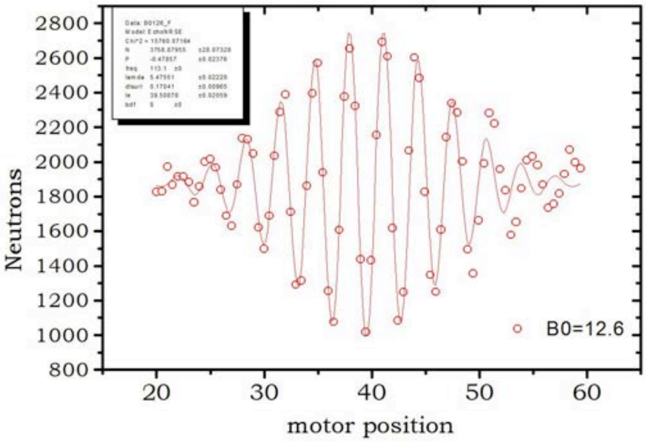


Fig. 9. The NSE signal measured with new coils shown in Figs. 7 and 8.

Polarized neutron technique for measurements of triplet correlation functions.

In the present day neutron experiment one determines the mutual position of two scattering objects in the sample: the distance between these objects is the length scale at which we probe the sample (so-called pair correlation function. However, using the NRSE technique one can split the initial neutron wave into four waves, so that in the precession path of the first NSE arm these four neutron waves will produce phase shifts corresponding to three different distances: these neutron waves probe the sample simultaneously on three length scales. The detected intensity will now depend on distances between three scattering objects in the sample (so-called triplet correlation function). The prototype setups for the Four-Wave Neutron Resonance Spin Echo (FW-NRSE) experiments and Spin-Echo Small Angle Neutron Scattering (SESANS) has been built at the reactor of PNPI, in Gatchina, Russia.

Ultra-Small-Angle Polarized Neutron Scattering (USANSPOL)

The characterization of magnetic structure of matter and the study of its evolution in varying external conditions is a particular strength of neutron research. A new option for investigating the magnetic microstructure is the USANSPOL technique, which relies on the high resolution of scattering angles provided by perfect crystal neutron reflection. A corresponding instrumental configuration was set up at the S18 instrument at the ILL, Grenoble (Fig.10-11). This instrument was equipped with magnetic prisms for neutron polarization and a dedicated sample environment, which provides 3D control of an external magnetic field configuration ranging from a zero-field environment via continuous intermediate settings up to the magnetic saturation of the samples. In addition, this sample environment enables us to apply external mechanical forces for the study of magnetostriction in novel technologically relevant materials that are used in modern sensors and actuators. Exemplifying test experiments were performed which demonstrated the potential of the new technique (Fig.12). Measurement results allow for an assessment of the native sample state

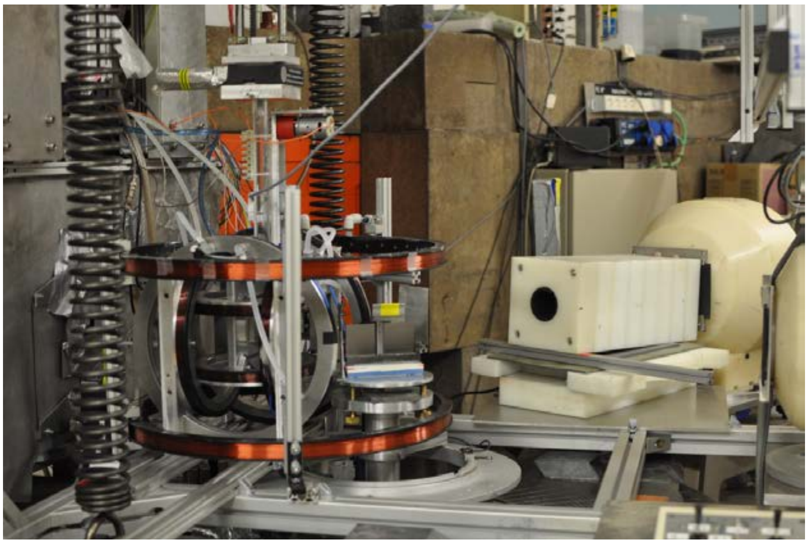


Fig.10. Implementation of the USANSPOL setup at the instrument S18 at ILL, Grenoble. Neutrons enter the instrument from the left where the monochromator crystal and the prism polarizers are located. The dedicated USANSPOL sample environment is shown in the left part, the analyzer crystal in the centre and the detector for the scattered neutrons in the right part of the picture. The sample environment provides 3D control of the external magnetic field at the sample position. The sample itself is mounted on a sample holder, which can subject it to mechanical stress, exerted by the torque of a motor which can be seen in the upper central part of the image. The large pair of Helmholtz coils generates a magnetic guide field in the sample/analyzer crystal area.

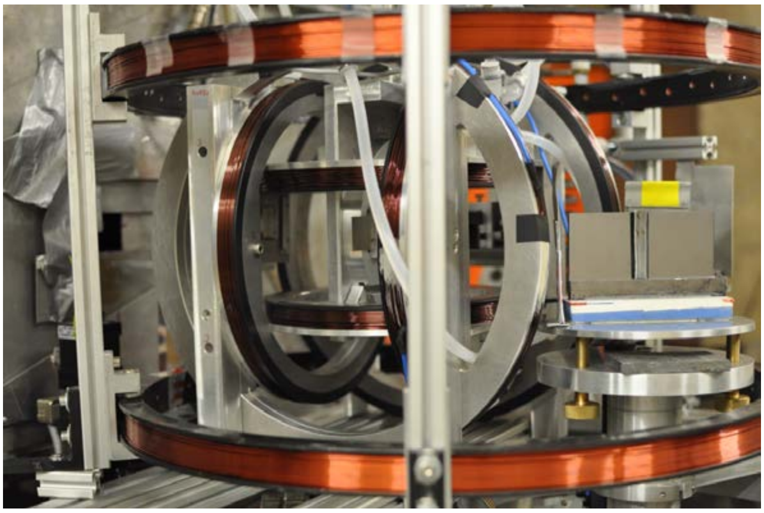


Fig. 11. The image offers a detailed view of the USANSPOL sample/analyzer crystal area. The large pair of Helmholtz coils generates a magnetic guide field to define the initial neutron beam polarization. The 3 pairs of coils in the centre of the image enable a 3D control of the external

magnetic field at the sample position. The analyzer crystal of the double crystal diffractometer is shown in the right part of the picture.

which follows from a specific manufacturing process and the evolution of its microstructure in external magnetic fields and under mechanical stress according to its particular magnetic properties. Thereby, USANSPOL experiments offer a picture of the structure which may be related to material functionality and eventually lead to technological improvement.

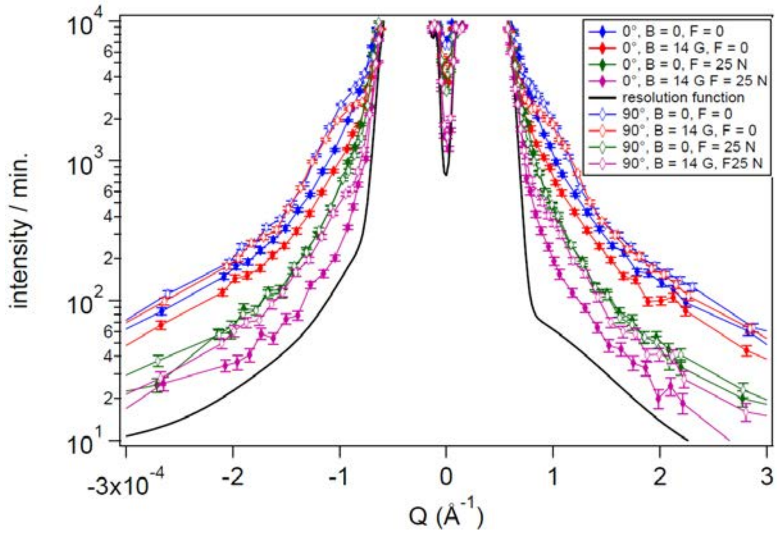


Fig. 12. Experimental results of USANSPOL measurements of a magnetic ribbon ($Fe_{78}Mo_2B_{20}$). These measurements were performed with horizontal (0°) and vertical (90°) ribbon orientation. The samples were placed in magnetic zero-field (B = 0) and an external magnetic field of B = 14 G. Additionally a mechanical force of 25 N was applied in the various configurations.

Development of an ultra-flexible neutron magnetic resonator

Spectral and temporal tailoring of neutron beams is an important issue for advanced neutron sources like the European Spallation Source project. Spatial magnetic neutron spin resonance as basis for the famous Drabkin-resonator demonstrated its potential for defining the spectrum of a polarized neutron beam already in the 1960s. With the novel idea of controlling each element of the neutron resonator separately, a concept was invented that allows for polarized neutron beam tailoring of unprecedented flexibility regarding key parameters like incident and final neutron energy, spectral width of the incoming beam, or its energy resolution. We have built two prototype resonators according to that concept and tested them experimentally at a polarized neutron beam line at the Atominstitut TRIGA reactor of the Vienna University of Technology (Fig.13-14). The experiments demonstrated the flexible spectral definition of the neutron beam with variable resolution in continuously operated mode which may be applied in diffraction and fundamental physics experiments.
They also included the first realization of a travelling wave mode of operation where short neutron pulses in the microsecond range are produced by synchronized magnetic field pulses. This operation mode offers new possibilities for neutron spectroscopy as it decouples the energy resolution from the time-of-flight resolution of the neutron beam. In addition, a Ramsey-type resonator setup was conceived and also tested which offers promising potential for advanced neutron spectroscopy techniques.

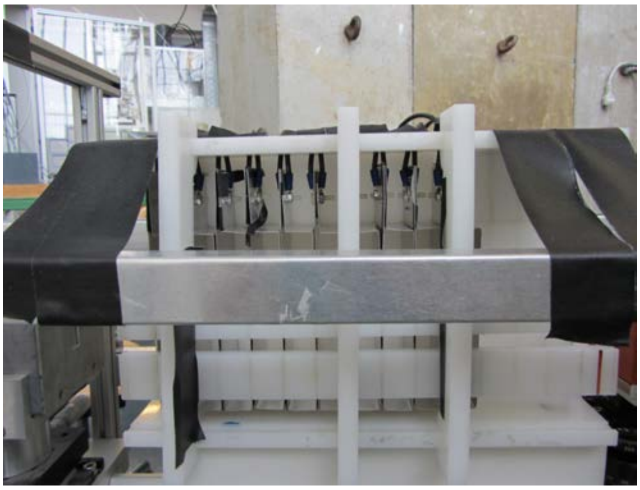


Figure 13. Prototype 1 of a travelling-wave mode spatial magnetic neutron spin resonator consisting of 10 individually tune- and switchable aluminum coils. The aluminum profile in the centre of the picture accommodates the magnetic coil which produces the magnetic selector field which define the resonance wavelength for the neutron beam.

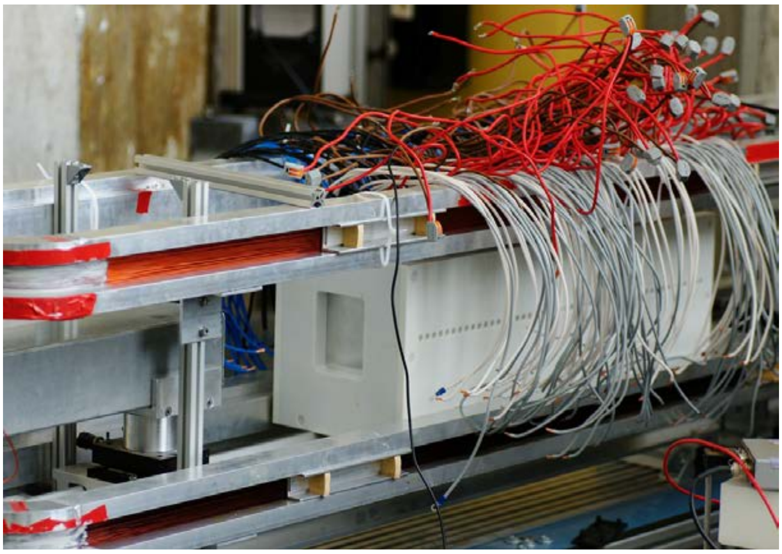


Fig. 14. Side view of the prototype 2 spatial magnetic spin neutron resonator. The device consists of 48 novel triple-stage resonator elements with 288 connector cable in total. The resonator is placed inside the coils which generate the magnetic selector field. In the left part of the image the entrance part of the supermirror analyzer can be seen.

Conclusions.

In the past, neutron polarization analysis experiments were considered as delicate, complicate for understanding and often costing too much flux. Decisive progress was made recently in instrumentation for and understanding of polarized neutron scattering provides vastly increased new power for delicate experiments investigating the nature of magnetism and magnetic phenomena in solids. Polarized neutron scattering is successfully developing in both of these directions and a number of remarkable results have been achieved. Developments made in the frame of the current project are obviously extending the power of polarized neutron scattering and showing the way forward.

Cooperative efforts of partners have been concentrated both on the use of polarised neutrons as an elementary magnetic probe in condensed matter investigations and the application of the Larmor precession (LP) technique for development of a new generation of LP-based neutron scattering instruments, methods and devices. This allowed for further improvements in the use of polarized neutrons in neutron scattering as the wide-angle polarization analysis and in construction of new generation of neutron scattering instruments using polarized neutrons (high-resolution spectrometers, diffractometers and reflectometers), as well as futher developments of the simulation techniques. New approaches using polarized neutrons, such as measurements of triplet correlation functions and development of an ultra-flexible neutron magnetic resonator have been successfully explored.