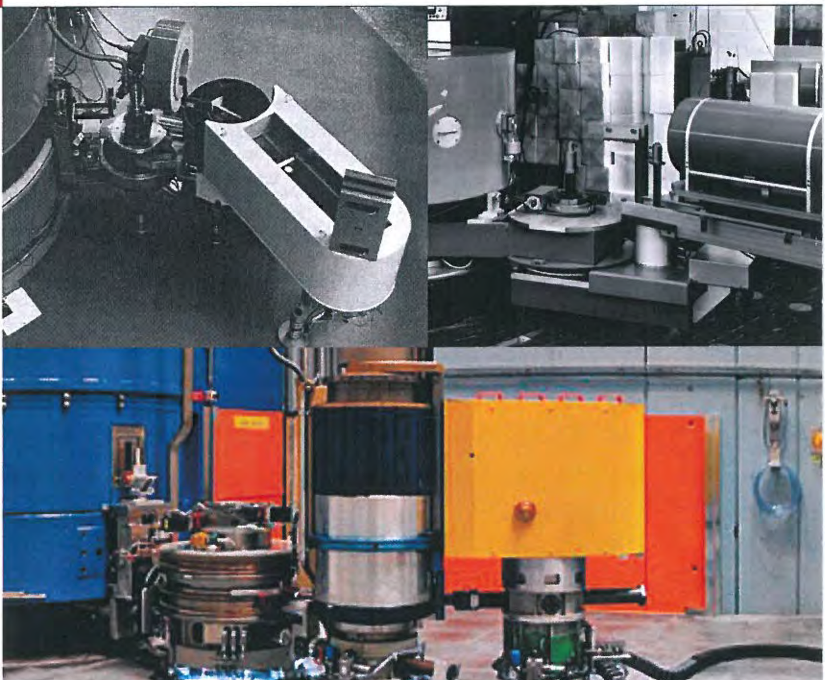
Number 43
March 2014

SWISS NEUTRON NEWS



Schweizerische Gesellschaft für Neutronenstreuung
Société Suisse pour la Diffusion des Neutrons
Swiss Neutron Scattering Society

On the History of Neutron Spectrometers in Switzerland

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The last issue of Swiss Neutron News (No. 42, August 2013) featured an article entitled *50 Years of Swiss Neutron Diffraction Instruments* by Peter Fischer *et al.*, which is complemented here by a chronological summary of the development of neutron spectrometers (i.e., instruments for inelastic neutron scattering) in Switzerland from the early days up to the present. This article is dedicated to the memory of Walter Hälg (1919–2011), the founder of neutron scattering in Switzerland.

HOW IT STARTED

The first Swiss neutron source was the light-water reactor *Saphir* which started operation in the year 1957 with a thermal power of 1 MW, but the thermal neutron flux of about 10^{13} n·cm^{-2}·s^{-1} was not sufficient for neutron spectroscopic experiments. The situation was improved in the year 1960 with the commissioning of the heavy-water reactor *Diorit* (30 MW thermal power) providing a thermal neutron flux of the order of 10^{14} n·cm^{2}·s^{-1}. Initially, inelastic neutron scattering experiments were performed with use of a **rotating-crystal time-of-flight spectrometer**, which was transferred in the year 1961 from the Kernforschungszentrum Karlsruhe (Germany) to the reactor *Diorit* by Wolfgang Gläser's group (because of a several years' shutdown of the Karlsruhe reactor). It was left at the disposal of Walter Hälg's group after the restart of the Karlsruhe reactor. The instrument was equipped with a rotating Al monochromator and a series of scintillation detectors. Not much was known at that time about the resolution properties of neutron spectrometers, so that the necessary knowledge had to be acquired by „learning on the job" [1,2]. The spectrometer was completely rebuilt in the year 1967 with improved resolution, intensity and background by using rotating Pb monochromators, He flight paths and He detectors (Fig. 1).

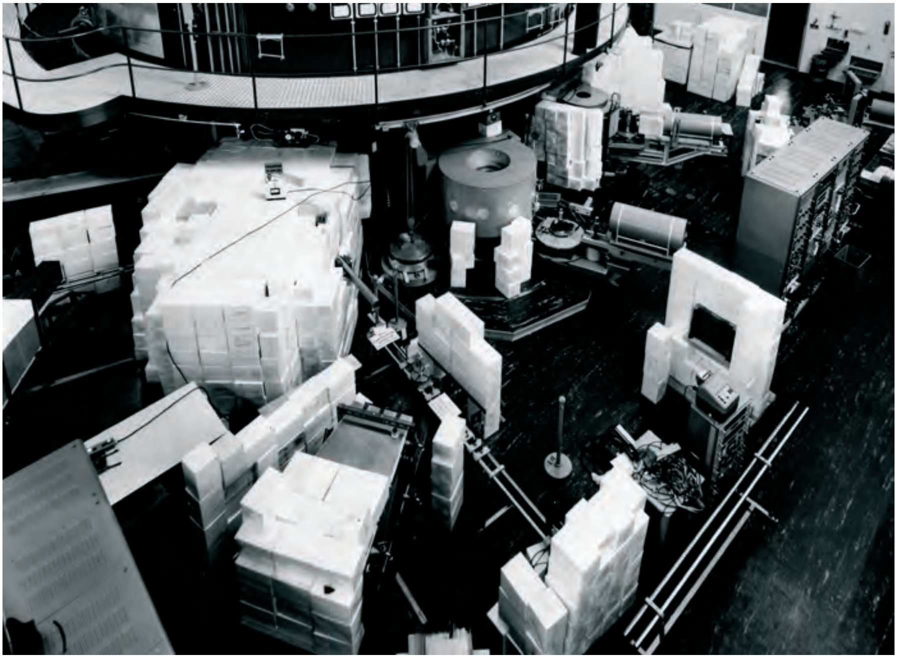

Figure 1: Instruments at Diorit in the year 1967. The rotating-crystal time-of-flight spectrometer is in the foreground. The neighboring instrument is the three-axis spectrometer.

AN INSTRUMENT BASED ON AN ANTI-AIRCRAFT-GUN CARRIAGE

The construction of a **three-axis spectrometer** started in the year 1963, and its installation at the reactor *Diorit* was completed in 1966. The monochromator axis was built up on top of a rotating anti-aircraft-gun carriage provided by the Swiss army (Fig. 2). Both the sample and the analyzer axes were carried along mechanically in a self-supporting manner and driven by cables, which resulted in severe problems due to rather long wiggling times around the final angular positions. Some years later, the „crazy" cable drives were replaced by a system of tooth wheels.

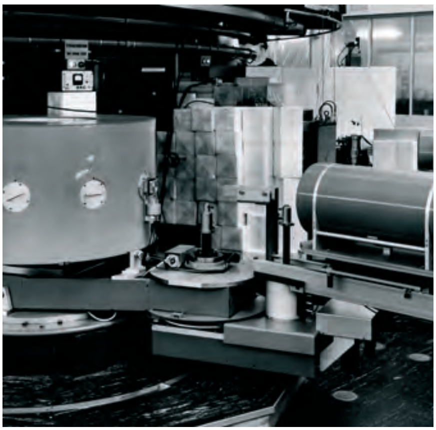

Figure 2: Three-axis spectrometer at Diorit. The monochromator is built on top of a rotating anti-aircraft-gun carriage.

The constant-Q or constant-ω scans had to be programmed on a central computer and fed to the instrument in the form of paper tapes. In order to run the spectrometer over the weekend, several hundred meters of paper tape had to be provided in advance!

THE FIRST STEP INTO POLARIZED NEUTRONS

In 1964 I stepped into neutron scattering to carry out the diploma thesis. My task was to produce **polarized neutrons** and to utilize them for spin-wave measurements. Large fcc $Co_{0.92}Fe_{0.08}$ plates mounted in permanent magnets served as both polarizer and analyzer crystals, and high-frequency spin-flip coils placed inside the guide field were used as π-inverters of the neutron spin. In order to achieve spin turnings by π/2, a spiral-type guide field was manufactured in which the neutron spin adiabatically followed the external magnetic field (Fig. 3). This device is now the most beautiful trophy in my office! The polarized neutron instrument was missing the third axis for energy selection, so that information on the spin waves could only be obtained by measuring the scattering surface away from the Bragg position, a method developed by Elliott and Lowde at the Harwell reactor in the U.K. [Proc. Roy. Soc. 230, 46 (1955)].

MOVING FORTH AND BACK BETWEEN DIORIT AND SAPHIR

In 1970 the reactor *Diorit* was shut down, because the reactor tank had to be replaced. Both the three-axis spectrometer and the rotating-crystal time-of-flight spectrometer were moved to the reactor *Saphir*, whose thermal power was increased to 5 MW in order to make neutron spectroscopic experiments possible. In 1973 the reactor *Diorit* was operational again, and the two spectrometers were moved back from the reactor *Saphir*.

ENTERING THE GRAPHITE AGE

At about the same time the high-flux reactor at the Institut Laue-Langevin (ILL) in Grenoble started to produce neutrons with a thermal flux of about 10^{15} $n{\cdot}cm^{2}{\cdot}s^{-1}$, *i.e.*, an order of magnitude larger than that of the reactor

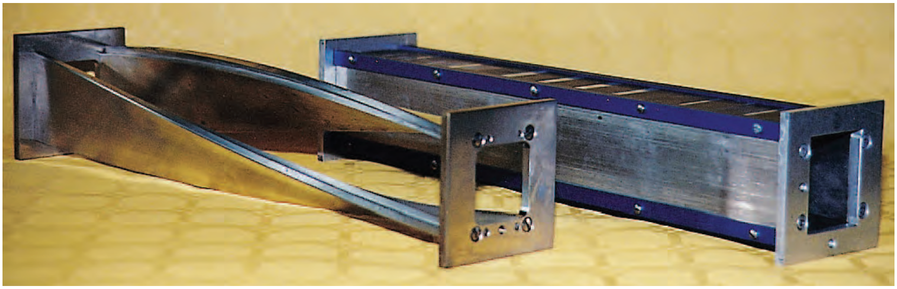

Figure 3: Magnetic guide fields

Diorit. In order to be competitive on an international level, many efforts were undertaken to optimize the flux conditions at the Swiss neutron spectrometers. Motivated by a paper of Tormod Riste (1925–1995) [Nucl. Instrum. Meth. 75, 197 (1969)], a **pyrolitic graphite** plate with an area of 5x5 cm^2 was purchased in 1970 and inserted as monochromator for the three-axis spectrometer, which resulted in an intensity increase by factors between 2 and 5. Some years later vertically bent graphite crystals (with fixed bending) became available, which roughly doubled the intensity without losing resolution. At that time oriented graphite crystals were extremely expensive, contributed to an „explosion" of the laboratory budget and therefore had to be treated very carefully; nevertheless, Willi Bührer (1938–1997) once took the liberty of signing an order with use of the corner of a graphite crystal instead of an ordinary pencil!

THE FIRST „EXOTIC" SPECTROMETER

Another idea realized at the reactor *Diorit* in 1973 was the **MARC** (**M**ulti-**A**ngle **R**eflecting **C**rystal) **spectrometer** based on an instrument concept developed at Risø National Laboratory (Denmark) by Jørgen Kjems [*Neutron Inelastic Scattering*, IAEA, Vienna (1972), p. 733]. In principle, the MARC spectrometer is a conventional three-axis spectrometer, but it differs from the latter by the analyzer and detector system (Fig. 4). The neutrons scattered from the sample are collected over a large range of scattering angles and then energy selected by a large analyzer crystal (with a mosaic spread of several degrees) and by a position- sensitive detector, which allows the simultaneous measurement of a complete energy spectrum. Due to a delay in the provision of the detector electronics, the MARC spectrometer was initially used as a three-axis spectrometer, and only in 1976 it was operated in the desired MARC scheme, thereby demonstrating the expected overall intensity gain by an order of magnitude. The MARC spectrometer was controlled by a PDP 11/10 computer and CAMAC electronics, and the heavy weights were moving on homemade air cushions. At the same time, this „modern" instrument technology was also applied to completely upgrade the „old-fashioned" three-axis spectrometer.

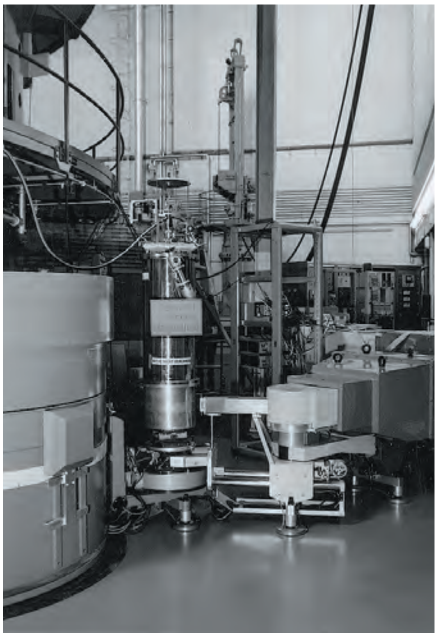

Figure 4: The MARC spectrometer at Diorit

A DEVICE INSPIRED BY THE GREEK MYTHOLOGY

Often single crystals of advanced materials can only be grown with volumes of a few mm^3, which contrasts to the usual size of neutron beams with cross sections in the cm^2 range. In order to make better use of the neutrons at the sample position, our skilled technicians realized a multi-crystal goniometer in the year 1974, following an idea of Bill Buyers (Chalk River National Laboratory, Canada). Up to seven single crystals could be individually adjusted (Fig. 5). The system was manufactured by brass in a rather compact way, suitable for insertion into cryostats. The **multi-crystal goniometer** was called **Hydra**, referring to the multi-headed snake-like monster in the Greek mythology.

Figure 5: The multi-crystal goniometer Hydra

MOVING AGAIN FROM DIORIT TO SAPHIR

1977 marked the year of the final shutdown of the reactor *Diorit*. In the meantime the experimental hall of the reactor *Saphir* was considerably enlarged in order to provide sufficient room for the neutron scattering activities. However, only four radial beam ports were available, two for neutron diffraction and two for neutron spectroscopy. Therefore only the very successful three-axis and MARC spectrometers could be reinstalled at the reactor *Saphir*, whereas the rotating-crystal time-of-flight spectrometer had to share its fate with the reactor *Diorit*.

A RECIPE TO SURVIVE WITH MODERATE FLUX

Due to the limited flux of the reactor *Saphir* the efforts to optimize the neutron spectrometers were continued. In particular, **the idea of horizontal focusing** proposed at the ILL Grenoble by Reinhard Scherm [Nucl. Instrum. Meth. 143, 77 (1977)] was brought to Swiss perfection. A very flexible system called **Jalousie** (= Venetian blind) was realized (Fig. 6), which was composed of seven individually adjustable goniometer heads [3]. The latter were mechanically coupled to achieve rotations of the outer crystals by $\pm\delta$, $\pm\delta 2$, $\pm\delta 3$ with respect to the central one. The three-axis spectrometer was upgraded in the year 1980 with two *Jalousie* systems: For the monochromator high quality and vertically bent graphite crystals were used and for the

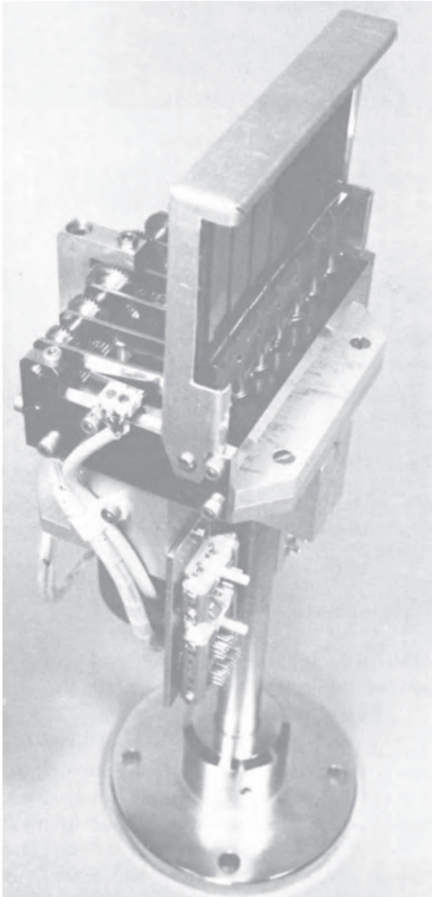

Figure 6: Jalousie monochromator with seven graphite crystals

analyzer medium quality flat ones. An intensity gain factor of the order of 20 resulted from this upgrade without losing energy resolution. In fact, the use of this kind of beam focusing was extremely successful, so that some years later also the MARC spectrometer was transformed into a three-axis spectrometer with a doubly focusing monochromator and a horizontally focusing analyzer.

TO BELIEVE, OR NOT TO BELIEVE, THAT IS THE QUESTION

The international neutron scattering community expressed a tremendous interest in our focusing monochromator and analyzer systems. However, some people were not really enthusiastic about beam focusing. I remember a visit of Gen Shirane (1924–2005), the neutron scattering „guru“ of Brookhaven National Laboratory, in the early nineties. He doubted that focusing systems can be operated reliably on a long time scale. In particular, he did not believe that after some hundred thousand measurement scans the angular settings of the individual crystals can be reproduced exactly. Obviously he underestimated the mechanical skill of Swiss technicians! After Gen Shirane's visit we used to categorize three-axis experiments into either „Brookhaven type“ (no focusing, low intensity, poor statistics) or „Swiss type“ (full focusing, high intensity, excellent statistics).

SQUEEZING MORE NEUTRONS OUT OF SAPHIR

In 1983 the thermal power of the reactor *Saphir* was upgraded from 5 MW to 10 MW. At the same time the dimensions of the reactor beam tubes were doubled from 4×4 cm^2 to 8×4 cm^2 and filled with helium (to avoid intensity loss through scattering from air) [4]. In addition, silicon filters cooled to liquid nitrogen temperature were inserted into the beam tubes to minimize the background. All these measures resulted in an overall intensity gain of the order of 5, which was extremely beneficial for neutron spectroscopic experiments. In fact, the three- axis spectrometers at *Saphir* were fully competitive with those at neutron sources with higher flux. Moreover, for particularly demanding experiments, the head of the reactor *Saphir* sometimes agreed to run the source for a couple of hours beyond the legal limit of 10 MW at a power of 12 MW. Normally a bottle of wine for the reactor crew was the minimum fee for this favor!

SAMPLE ENVIRONMENT: A KEY FOR SUCCESS

Permanent efforts were undertaken to improve the performance of the three-axis spectrometers as well as to extend the sample environment by novel devices. In collaboration with the Nobel Prize winner Alex Müller a uniaxial pressure device for insertion into cryostats and pressures up to 1 Mbar/mm^2 was constructed [5]. For optically active materials a device was developed for performing neutron scattering experiments with simultaneous irradiation by light pulses at low temperatures [6]. In 1992 a very compact analyzer-detector system was taken into operation, which allowed optimal horizontal focusing by properly adjusting both the position and the aperture of the detector (Fig. 7). Moreover, the shielding was optimized such that no enhancement of the background was measured when the detector was moving through the monochromatic beam.

THE SWISS EXILE AT THE ILL GRENOBLE

The „neutron paradise“ at *Saphir* did not last forever. In 1992 the thermal power of *Saphir* was reduced to 5 MW, and at the end of 1993 the reactor had to be shut down, mainly for safety reasons, but also in view of the new spallation source *SINQ* which was already in an advanced planning stage. Fortunately, in the early nineties the ILL Grenoble was looking for Collaborating Research Groups (CRG) to operate some of the existing instruments. In 1994 the ILL and the Paul Scherrer Institut (PSI) signed a contract to use the three-axis instrument IN3 in the framework of a CRG to bridge temporarily the neutron gap in Switzerland (Fig. 8). The flux at IN3 (installed at a thermal neutron guide) was rather moderate, but the insertion of a horizontally focusing trumpet between the monochromator and the sample resulted in a significant intensity gain. The CRG operation was terminated in 1998.

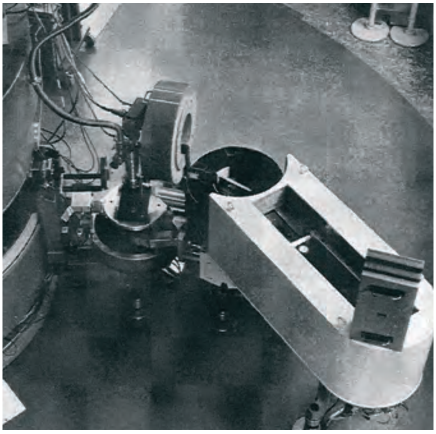

Figure 7: Compact analyzer-detector system of the three-axis spectrometer at Saphir

ENTERING A NEW AGE WITH SINQ

The spallation neutron source *SINQ* produced the first neutrons on December 3, 1996, followed by a commissioning period for the neutron scattering instruments, which are controlled by a standardized electronics and software system [7]. Scheduled user operation started in 1998 with a proton current of 1.6 mA. The first target made of Zircaloy and cooled by heavy water produced a thermal neutron flux of $n=0.6\times10^{14}$ $n\cdot cm^{-2}\cdot s^{-1}\cdot mA^{-1}$. In the year 2000, the target was replaced by a system of lead rods in Zircaloy tubes, which doubled the thermal neutron flux. At the same time, the proton current was increased to 2.0 mA, resulting in a thermal neutron flux of $n=2.4\times10^{14}$ $n\cdot cm^{-2}\cdot s^{-1}$ typical of a medium-flux neutron source.

THREE-AXIS SPECTROMETERS, OF COURSE!

For the first generation of neutron spectrometers, emphasis was led on the use of cold neutrons because of the very efficient cold neutron source sitting close to the target. The **three-axis spectrometer DrüchaL** [**Drü**achsigs am **cha**lte **L**eiter] installed at the cold neutron guide No. 13 (Fig. 9) was ready for inelastic neutron scattering experiments in early 1997, followed some months later by the **three-axis spectrometer TASP**, which mechanically is a copy of DrüchaL, but it has the option to make use of polarized neutrons. Both DrüchaL and TASP are moving on air cushions on a sturdy and maintenance-free granite floor. TASP is located at the end of the cold neutron guide No. 14, thus it can make

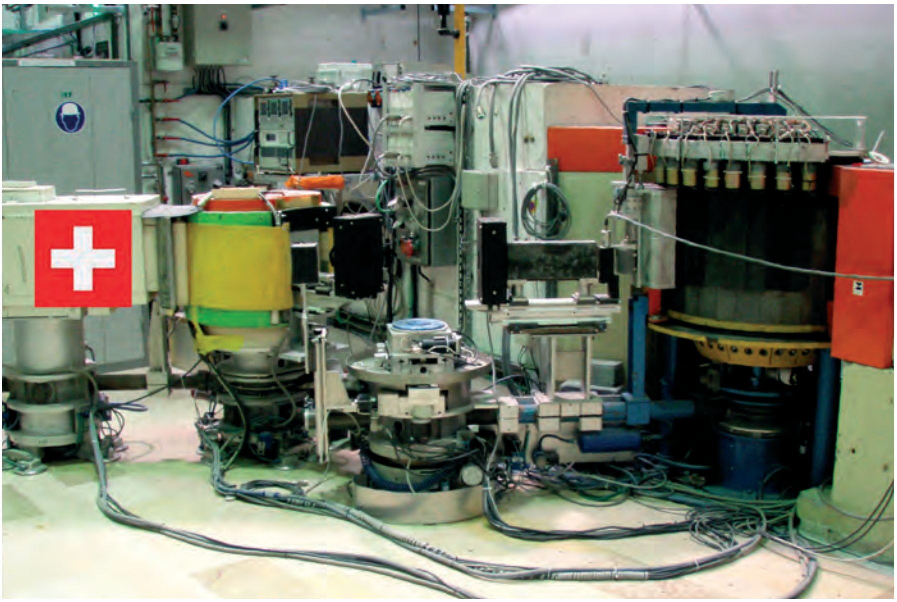

Figure 8: The „Swiss" three-axis spectrometer IN3 at the ILL Grenoble

full advantage of this privileged position by employing collimation or using a Heusler monochromator in the primary beam. An important addition to TASP was made in the year 2008 by the availability of **MuPAD**, which is a **Mu**-metal **P**olarization **A**nalysis **D**evice allowing the arbitrary orientation of a polarized neutron beam, so that spherical neutron polarimetry and xyz polarization experiments can be performed.

TOWARDS HIGH RESOLUTION

The **high-resolution time-of-flight spectrometer FOCUS**, developed in collaboration with Rolf Hempelmann (University of Saarbrücken, Germany) and partly funded by the BMBF (Bundesministerium für Bildung und Forschung, Germany), became operational in 1999 (Fig. 10). FOCUS, located at the end of the cold neutron guide No. 11, is a hybrid crystal-chopper spectrometer with either monochromatic or time focusing option [8,9]. The primary spectrometer consists of three parts. The white incoming neutron beam, reduced in size by a converging guide, is cut into packets by a disk chopper. A Be-filter can be inserted after the disk chopper to suppress higher-order contamination. The neutron packets are then deflected by a horizontally and vertically focusing monochromator equiped with either pyrolitic graphite or MICA [10] crystals, before they are chopped again in a Fermi chopper. The secondary spectrometer consists of detector banks surrounding the sample from +10° to +130°. The initial number of 200 He detectors of rectangular shape was later increased to 375, and a two-dimensional multidetector was added to cover the angular range from -5° to -24°. An oscillating large-angle collimator is inserted between the sample and the detectors in order to avoid scattering from the cryostat. FOCUS opened the way to neutron spectroscopic experiments with energy resolutions down to 10 μeV.

Figure 9: The three-axis spectrometer DrüchaL at SINQ with Willi Bührer (left) and Peter Böni

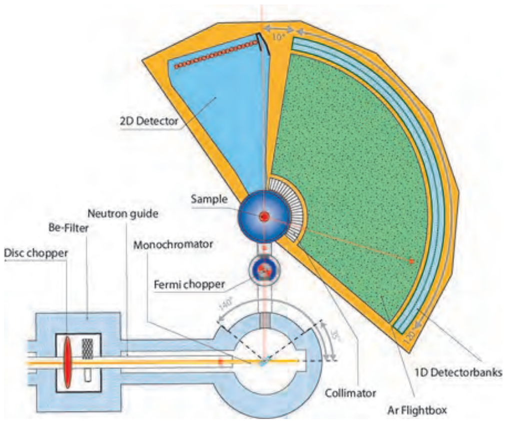

Figure 10: Sketch of the high-resolution time-of-flight spectrometer FOCUS at SINQ

EN VARM VELKOMST HILSEN VED RISØ!

In the year 2001 the Danish neutron scattering group moved its experimental activities from Risø National Laboratory to SINQ. This had consequences for the three-axis spectrometer DrüchaL, whose analyzer and detector parts were exchanged against the **RITA** (**R**e-**I**nvented **T**hree-**A**xis Spectrometer) system developed at Risø. As a result, DrüchaL was renamed as RITA-II (Fig. 11). RITA-II allows very flexible configurations due to the use of a multi- blade analyzer and a position-sensitive detector, but it can also be used in the conventional three-axis mode. A particularly attractive configuration is the monochromatic q-dispersive mode, which provides a mapping in the energy-wavevector space.

HIGH RESOLUTION AT ITS BEST

The second high-resolution neutron spectrometer realized at SINQ is **MARS** (**M**ulti-**A**ngle **R**eflecting Crystal **S**pectrometer) [11], an inverted time-of-flight backscattering spectrometer similar to IRIS at the spallation source ISIS (U.K.). MARS is situated at the cold neutron guide No. 15 in an extension of the guide hall finished in 2004, and user operation started in 2007. Five choppers define the narrow energy range of the incoming neutrons. The secondary spectrometer merges alternating diffraction and inelastic units, and the final neutron energy is determined by reflection from five large, moveable analyzer banks equiped with MICA crystals. The corresponding ^{3}He detector tubes are moveable as well around the near backscattering position (Fig. 12). MARS boasts high resolution over a large

Figure 11: The three-axis spectrometer RITA-II at SINQ

energy range, reaching 1 μeV at the elastic line. Unfortunately, MARS was shut down in 2013 for several reasons.

CLIMB UP THE EIGER!

The youngest member of neutron spectrometers at SINQ is the three-axis spectrometer **EIGER** (**E**nhanced **I**ntensity, **G**reater **E**nergy **R**ange) utilizing thermal neutrons, thereby expanding the available energy range for neutron spectroscopy in a substantial manner. EIGER started user operation in the year 2011 (Fig. 13). It is situated at the thermal beam port No. 82 equipped with a sapphire filter (to suppress neutrons with energy >80 meV) and a focusing guide, which is beneficial for both high flux and low background. A particular feature is the monochromator shielding built from non-magnetic materials, so that high-field cryomagnets can be used.

WHAT CAN BE LEARNED?

Looking back to fifty years of activities in the development of neutron spectrometers in Switzerland, I come to two major conclusions:

1. The time needed for building a new spectrometer, i.e., the time from the idea to the final realization, has been increasing substantially. I became aware of the fascinating MARC principle during a visit at Risø in late 1970, and already in early 1973 we had a MARC spectrometer operational at the reactor *Diorit*. On the other hand, the feasibility study for MARS was completed in 1997, and in the same year an international expert committee gave a strong recommendation to realize MARS, but the project was completed only in 2007! Obviously much time is needed today to overcome all the „hurdles" (technical feasibility study, scientific case, reports by experts, financial planning and sponsorship, availablity of technical manpower)

Figure 12: Analyzer and detector banks of the backscattering spectrometer MARS at SINQ

towards the final commissioning of the instrument.

2. The well known saying **„every neutron is a good neutron"** has been repeatedly verified. Inelastic neutron scattering at medium-flux neutron sources like *Saphir*, *Diorit* and SINQ can be absolutely competitive with the conditions offered at high-flux neutron sources, provided that clever people are around who persistently try to incorporate innovative ideas into the spectrometers, with the aim to transport as many (useful) neutrons as possible to the detector.

PREDICTING THE FUTURE?

In order to maintain as well as to extend the present level of neutron spectroscopy in Switzerland, instrument improvements and novel possibilities should be permanently envisaged:

1. **The SINQ neutron guides**, designed and realized twenty years ago as the most efficient guide system at that time, urgently need an upgrade now. Novel ballistic guides are able to increase the flux at many instrument positions by almost an order of magnitude [12].
2. **Focusing** some neutron beams down to the mm^2 range is another must to efficiently perform experiments on small samples (e.g. in high-pressure cells) [13].
3. The project **CAMEA** (**C**ontinuous **A**ngle **M**ultiple **E**nergy **A**nalysis) constitutes the ultimate step from TAS via MARC and RITA to a new generation of highly efficient three- axis spectrometers.
4. **Polarization devices** have proven to be most useful assets also for spectrometers [14].
5. Having the world's most powerful ultracold neutron source operational at PSI, the idea of **phase-space transformation** [15,16] might have a revival to deliver thermal neutrons by up-scattering, providing fluxes comparable to today's best thermal time-of-flight spectrometers, but with considerably better energy resolution.
6. Finally, and as a surprise, SINQ is still missing the **neutron spin-echo technique**!

Figure 13: The thermal three-axis spectrometer EIGER at SINQ

Ein Mann, der recht zu wirken denkt,
muss auf das beste Werkzeug halten

Johann Wolfgang von Goethe, Faust, Vorspiel (Direktor)

A bibliography of articles published with respect to the instrumental development of neutron spectrometers in Switzerland:

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