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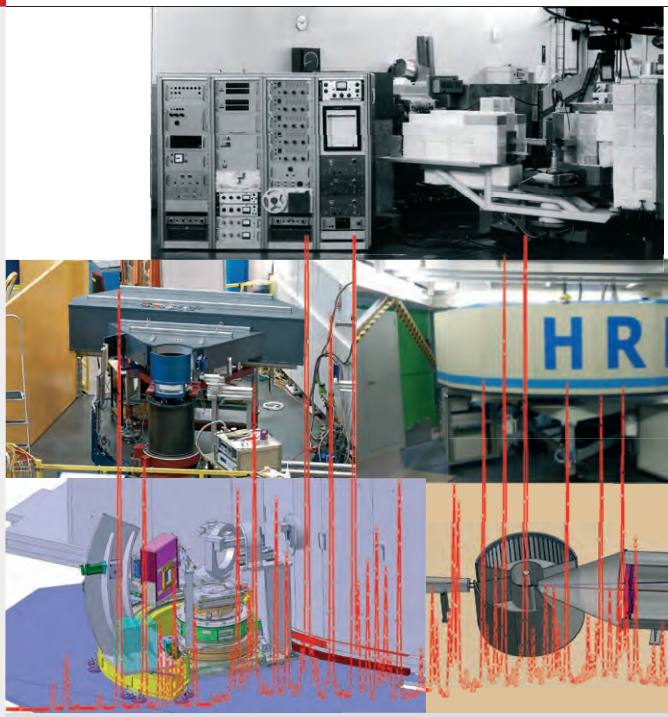


Schweizerische Gesellschaft für Neutronenstreuung
Société Suisse pour la Diffusion des Neutrons
Swiss Neutron Scattering Society

50 Years of Swiss Neutron Diffraction Instrumentation

P. Fischer [a], J. Schefer [b], L. Keller [c], O. Zaharko [d], V. Pomjakushin [e], D. Sheptyakov [f], N. Aliouane [g] and M. Frontzek [h]
Laboratory for Neutron Scattering, Paul Scherrer Institut, 5232 Villigen PSI, Switzerland
S. L. Holm [i] and K. Lefmann [j]
Nano-science Center, University of Copenhagen, Denmark
M. Christensen [k]
Department of Chemistry & iNano, University of Aarhus, Denmark

‡

1. INTRODUCTION [a, b]

Referring to the obituary for Prof. Walter Hälg (1917–2011) as Swiss neutron scattering pioneer [1], it may be worthwhile to look at the development of Swiss neutron diffraction instrumentation in the corresponding period of about 50 years.

It depended primarily on the neutron sources, their neutron beam channels, available space for the instruments and on the source operation modes and related political decisions.

First the light-water research reactor SAPHIR (Fig. 1) became critical in 1957 with a power of 1 MW. Depending on the experimental needs, the power had been increased in 1970 and 1983 to maximum values of 5 MW and 10 MW, respectively. In December 1993 occured its final shutdown.

In the year 1960 the Swiss heavy water reactor DIORIT I became critical. It reached maximum powers of 20 MW (neutron flux 3.5×10^{13} $ncm^{-2}s^{-1}$) and 30 MW in the years 1961 and 1966, respectively. In a long shutdown from 1970 to 1972 a new heavy water tank had to be installed.

Then DIORIT II operated until the final shutdown in 1977 with a maximum power of about 24 MW.

From 1988 on Switzerland has officially access to the neutron scattering instrumentation around the high flux reactor of the Institut Laue-Langevin (ILL) in Grenoble.

‡ Article posthumously dedicated to the Swiss neutron scattering pioneer Prof. Walter Hälg (1917–2011).

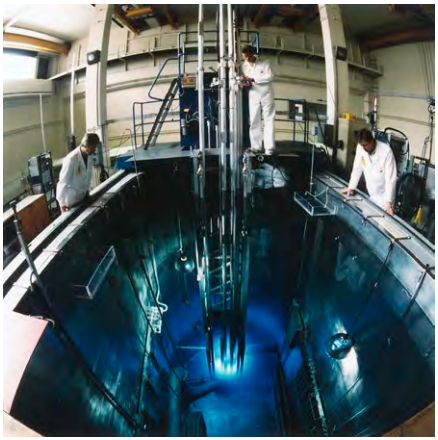


Figure 1: Characteristic Tscherenkow light of reactor SAPHIR (from picture archive PSI).

1994–1998 D1A at ILL could be also partially used in the CRG mode, see section 3.

In the year 1996 the continuous Swiss spallation neutron source SINQ started operation, a project based on ideas of Prof. W. Hälg and realized by a team under W. E. Fischer in cooperation with Prof. A. Furrer et al..

We shall restrict our review mainly on classical powder and single crystal neutron diffraction.

2. NEUTRON DIFFRACTOMETERS AT REACTORS SAPHIR AND DIORIT [a b]

During his stay 1952–1953 in Norway Prof. W. Hälg came at the reactor JEEP at Kjeller into contact with neutron scattering. Trained in optical spectroscopy, particle physics as well as in electronics from the University at Basel, he initiated at the swimming pool type reactor SAPHIR the construction of a first two-axes neutron diffractometer (Fig. 2).

Figure 2: Main mechanical parts of the first neutron diffractometer at reactor SAPHIR, supervised by the mechanics expert M. Koch.

As at that time electronic controls and computers were only in their beginnings, mechanical 2:1 coupling of the two axes could be used as option, based on the geometrical relations of the central and peripheral angles of a circle.

Such a measurement on a lead crystal, performed manually and noted in 1960 by W. Hälg, is shown in Fig. 3.

Dr. Georg Maier, a cousin of the German neutron scattering pioneer Prof. H. Maier-Leibnitz and Peter Fischer as thesis student were the first neutron scattering collaborators of W. Hälg. Fig. 4 shows a corresponding picture at the Swiss Federal Institute for Reactor Research (EIR), Würenlingen.

Together with W. Hälg we first tested neutron monochromators and started optimization of the instrument shielding (Fig. 5) that originally consisted mainly of boron-paraffin blocks and lead.

As the heavy water reactor DIORIT operated at considerably higher power than reactor SAPHIR, the two-axes neutron diffractom-

eter had been transferred to this reactor. Thus we got practical experience concerning the complementary aspects of thermal neutrons as particles and as waves and also tried to understand theoretically neutron reflectivity from monochromator crystals [2]. On the other hand G. Maier developed first programs to calculate neutron structure factors and for data evaluation at a Zuse computer of EIR.

Other necessary devices such as plugs, Soller collimators or crystal holders had been realized in collaboration with the workshop group under the supervision of W. Hälg and the workshop chief E. Härdi from EIR.

With respect to long measuring times automation of data collection had been necessary. Due to his electronics experience W. Hälg created soon a corresponding working group for this important project. A first result for the powder neutron diffractometer at reactor DIORIT is shown in Fig. 6.

Concerning the detector shielding W. Hälg proposed to test the possibility to position the detector accurately and to let the heavy shielding follow the detector movement. As for focusing the detector has to turn towards the monochromator shielding, later a more compact detector shielding such as shown in Fig. 7,

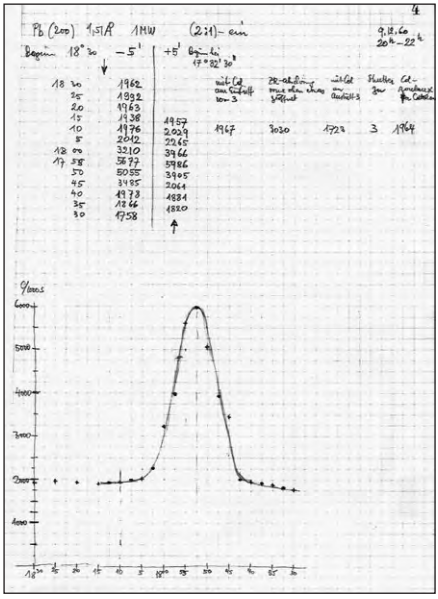


Figure 3: 1.5 Å neutron (200) intensity versus Bragg angle Θ of a Pb crystal, measured by W. Hälg in the Θ–2Θ mode at reactor SAPHIR at a power of 1 MW.

Figure 4: Research team of Prof. Hälg's Delegation AF, approximately 1962: from left W. Hälg, guest G. Ehret from Karlsruhe, P. Fischer, G. Maier, chemist F. Brandt and reactor engineer F. Ferroni.

Figure 5: Installation of the two-axes neutron diffractometer at reactor DIORIT.

combined with good monochromator shielding, resulted in considerably improved experimental conditions.

Such efforts yielded 1964 a first neutron powder diffraction publication, see Fig. 8.

Occasionally also Prof. Paul Scherrer (Fig. 9) passed by at the neutron diffractometer and checked wether the powder sample was properly rotating at room temperature according to the Debye-Scherrer method.

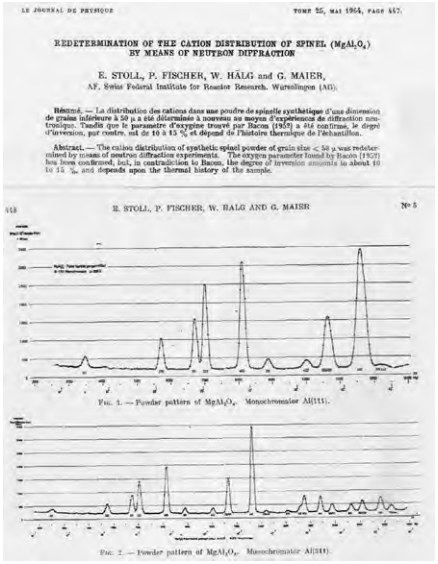
Fig. 10 illustrates the enlarged team of W. Hälg 1970 contributing to neutron scattering. Willi Bührer developed also with Swiss mechanical precision single and double focusing monochromator systems, in particular when suitable pyrolithic graphite became available.

Fig. 11 is an aerial view of both institutes EIR and SIN (Swiss Institute for Nuclear Research) in 1971. The large building left of the high chimney is the one of reactor DIORIT. And left, almost hidden by trees, one can see the building of reactor SAPHIR.

Because of the upgrading period 1970–1972 from reactor DIORIT I to II, reactor SAPHIR's power had been increased in the year 1970 to 5 MW. And a part of the neutron

Figure 6: Powder neutron diffractometer with automatic data collection by means of paper tape input and output at DIORIT I.

Figure 7: Optimized detector shielding at DIORIT I with optional counter tilting for single crystals.



LE JOURNAL DE PHYSIQUE TOME 25, MAI 1964, PAGE 447.

REDETERMINATION OF THE CATION DISTRIBUTION OF SPINEL ($MgAl_2O_4$) BY MEANS OF NEUTRON DIFFRACTION

E. STOLL, P. FISCHER, W. HÄLG and G. MAIER,
AF, Swiss Federal Institute for Reactor Research, Wuerenlingen (AG).

Résumé. — La distribution des cations dans une poudre de spinelle synthétique d'une dimension de grains inférieure à 50 μ a été déterminée à nouveau au moyen d'expériences de diffraction neutronique. Tandis que le paramètre d'oxygène trouvé par Bacon (1950) a été confirmé, le degré d'inversion, par contre, est de 10 à 15 % et dépend de l'histoire thermique de l'échantillon.

Abstract. — The cation distribution of synthetic spinel powder of grain size < 50 μ was redetermined by means of neutron diffraction experiments. The oxygen parameter found by Bacon (1950) has been confirmed, but, in contradiction to Bacon, the degree of inversion amounts to about 10 to 15 %, and depends upon the thermal history of the sample.

E. STOLL, P. FISCHER, W. HALG AND G. MAIER

FIG. 1. — Powder pattern of $MgAl_2O_4$. Monochromator Al(111).

FIG. 2. — Powder pattern of $MgAl_2O_4$. Monochromator Al(311).

Figure 8: Improved neutron powder diffraction resolution due to increase of the scattering angle of the monochromator.

Figure 9: Monument plate for Walter Boveri and Paul Scherrer stating the successful start of the first Swiss reactor DIORIT.

instrumentation including two-axis diffractometers had been installed there. Because of the space limitations with at maximum three beam tubes this had been a difficult time for the Swiss neutron community.

Fig. 12 shows the final neutron scattering instrumentation at reactor DIORIT II in the years 1972–1977, characterized by movement of heavy loads on air cushions.

At the right side of the central triple-axes neutron spectrometer one may recognize the two-axis neutron diffractometer, used for single crystal studies of magnetic phase diagrams in external magnetic fields up to 60 kG. With a vertically focusing pyrolithic graphite monochromator since 1974 remarkable intensity gains had been obtained.

On the left side in the front K. Tichy had installed together with Prof. J. Benes a first four-circle neutron diffractometer for single crystals, see e.g. ref. [3].

Figure 10: Group photo 1970 of the collaborators of W. Hälg (left) and EIR contributing to neutron scattering. Note the Swiss pioneers W. Bührer and A. Furrer for inelastic neutron scattering in the front row from right.

Figure 11: EIR and SIN 1971.

The data collection had been done by means of a central CDC 8090 computer.

Both at DIORIT and at SAPHIR helium gas recovery systems had reduced the costs for liquid helium essentially.

Due to the final shutdown of reactor DIORIT (to reduce costs) neutron instrumentation had been again transferred to reactor SAPHIR in an increased experimental hall, see Fig. 13. To improve the background conditions, also BeO elements had been installed in the reactor. In 1983 SAPHIR reached the maximum power of 10 MW. Thus the neutron intensity became approximately comparable to the one of DIORIT II with 24 MW.

Since 1975 to his retirement in 1984 W. Hälg had been the head of the neutron scattering group within his Institute for Reactor Technics (IRT) at ETHZ. He always made the neutron instrumentation available to a broad national and international user community and introduced a fair user system to distribute the beam time.

Figure 12: Neutron scattering instrumentation at reactor DIORIT II 1972–1977.

In addition he organized national discussion meetings in 1973, together with Prof. H. Gränicher as director of EIR. In 1978 he also presented in another discussion meeting with B. Sigg first ideas for a SINQ spallation neutron source.

At SAPHIR with H. Heer as coordinator each instrument had been controlled by means of a PDP 11 and later LSI 11 computer, thus permitting online data evaluation.

In order to increase with medium neutron flux substantially the performance of the powder diffractometer, W. Hälg et al. started as successful teamwork the realization of the double-axis multicounter neutron powder diffractometer DMC [4,5]. It is illustrated in Fig. 14. This project had been financially supported by several Swiss institutes. And after now almost 30 years of operation this instrument is still well demanded at SINQ, using cold neutrons, see section 4.

Also Prof. A. Furrer – succeeding Prof. W. Hälg in 1984 – actively promoted instrumental development as head of the Laboratory for Neutron Scattering, ETH Zurich. In particular he looked for important auxiliary equipment, such as a dilution refrigerator reaching 7 mK (Fig. 15).

Figure 13: Neutron instrumentation at reactor SAPHIR 1983 with two-axis neutron diffractometers visible in the center of the pictures a) and b).

Fig. 16 illustrates the neutron scattering group at that time.

Finally in Fig. 17 the new 4-circle neutron diffractometer of J. Schefer with closed-cycle cooling machine and single detector is shown.

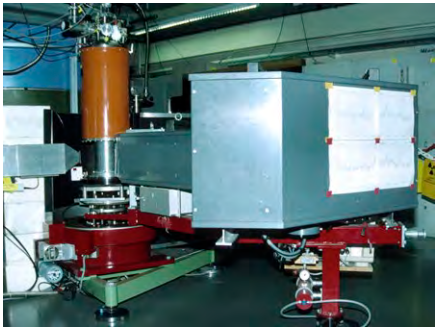


Figure 14: Final state 1993 of DMC with 400 BF3 detectors covering a scattering angle range of 79.8 degrees, radial collimator, optional 10′ mylar type primary collimator, vertically focussing pyrolithic graphite and Ge monochromators at the 10 MW reactor SAPHIR.

Figure 15: 7 mK refrigerator used 1988 on the two-axis neutron diffractometer P2AX@SAPHIR.

Figure 17: New four-circle neutron diffractometer 4C 1992 at reactor SAPHIR.

Figure 16: Neutron scattering group 1988.

3. D1A AS 'HALF SWISS' CRG INSTRUMENT [a]

From 1994 to 1998 Swiss users had between the shutdown of reactor SAPHIR and the startup of SINQ the opportunity to use up to 25 % of the D1A beam time at ILL in the CRG ('collaborative research group') mode for their research and for training of thesis students. In this period this first high-resolution powder neutron diffractometer of ILL [6] had 25 mylar type Soller collimators and ^{3}He detectors. And F. Fauth operated the instrument as local contact very well.

4. COLD NEUTRON POWDER DIFFRACTOMETER DMC AT SINQ [c h]

For the start of the Swiss Spallation Neutron Source SINQ in 1996 DMC was moved and adapted to the SINQ guide hall and has been operated since then as a cold neutron diffractometer. Located at an m = 2 supermirror neutron guide (Fig. 18), it is used without primary collimation and with optional secondary collimation providing maximum intensity. With the cold neutron spectrum (2.3 Å < λ < 5 Å), the focusing pyrolytic graphite monochromator and the low background due to optimized shielding, DMC is designed for efficient diffraction studies in the fields of crystallography, solid state physics and material science, in particular for the determination of weak intensities. Although its momentum transfer range Q is limited, its resolution exceeds the one of HRPT at smaller Q values. Special features are the linear position sensitive detector (BF_3, angular coverage 79.8°), the oscillating radial collimator system to suppress scattering from the sample environment and a large diversity of available sample environment devices, cf. http://www.psi.ch/sinq/dmc/.

A high-resolution option is provided by the optional vertically focusing Ge monochromator.

Designed complementary to the thermal instrument HRPT, typical experiments on DMC are the determination of magnetic structures, the efficient measurement of magnetic or crystallographic phase transitions, and the analysis of large unit cell structures.

Figure 18: High-intensity multidetector powder diffractometer DMC@SINQ for cold neutrons at SINQ.

Planned upgrades of the instrument include the replacement of the aging detector electronics and the installation of a non-magnetic sample table to further broaden the range of applications for DMC, in particular for investigations in external magnetic fields.

Standard for the control of SINQ instruments is the SICS client server system [7]. With it the instrument is locally supervised from the instrument computer, but measurements may

be also controlled remotely. And for online data evaluation either PC-s with Linux software or Mac-s are available.

5. HIGH-RESOLUTION POWDER DIFFRACTOMETER HRPT FOR THERMAL NEUTRONS AT SINQ [a e f]

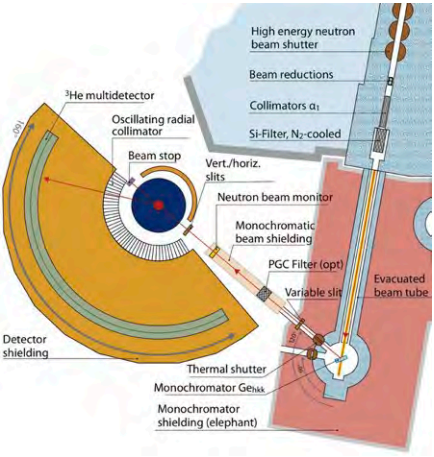


Figure 19: Layout of HRPT.

HRPT [8] is situated at the target station of SINQ (Figs. 19 and 20), using thermal neutrons from a water scatterer in a tangential beam-tube. Complementary to DMC it is designed as flexible instrument for both high-intensity and high-resolution investigations (see measured high-resolution functions shown in Fig. 21). In view of the medium neutron flux of SINQ and uncertainties at the beginning of SINQ operation concerning possible shielding problems due to the high energy spallation neutrons, this powder neutron diffractometer is based on a vertically focusing wafer-type Ge(hkk) monochromator, a radial collimator of mylar-Gd-O type and a large multidetector with 1600 wires and angular separation of 0.1 degrees between adjacent wires. The fixed monochromator takeoff-angles of 90 and 120 degrees ensure short monochromator-sample distances.

Figure 20: High-resolution powder diffractometer HRPT@SINQ for thermal neutrons with multidetector and electronics.

HRPT is designed as flexible instrument for efficient neutron powder diffraction studies in novel materials concerning chemical structures and magnetic ordering for large ranges of parameters such as temperature and pressure – also for small sample sizes. By means of a set of primary collimators, a secondary slit system and by appropriate choice of the sample diameter, resolution and intensity can be adapted to the needs, see http://www.psi.ch/sinq/hrpt/. The multidetector can be accurately positioned on air cushions.

The data transfer from the fast frontend field-programmable gate array FPGA, designed and programmed at PSI, to the user interface and histogram memory is made via a central data exchange system and optical cables.

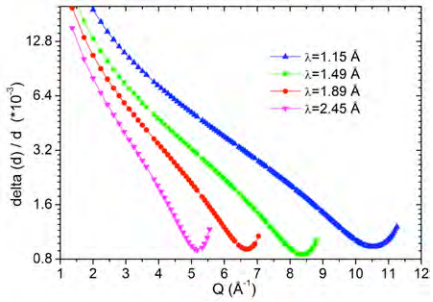


Figure 21: Measured high-resolution functions δd/d (α_1=6′, α_2=12′, radial collimator 2, sample diameter 6 mm) of HRPT for $2\theta_M$=120 degrees as functions of available neutron wavelengths and momentum transfer Q. d is the lattice spacing.

Figure 22: Platform on top of HRPT.

The high number of channels and high electrical voltage (~7 kV) provoke continuous occurrence of sporadic discharges that can lead to the appearance of false counts ("spikes"). Several hardware and software filters are implemented. A blocking trigger installed in FPGA filters these "discharges" by making use of their synchronous appearance in several non-neighbouring wires. The critical high voltage sockets are now continuously flushed with nitrogen gas. The detector is very well shielded also from the fast SINQ neutrons. As a result of all the above efforts the background conditions are very good, allowing measurements of rather small samples.

In the last decade important new auxiliary devices/possibilities such as a platform for convenient sample handling, cooling liquids etc., were added. It is illustrated in Figs. 22a) and b).

One may now choose between two oscillating radial collimators (FWHM = 7 mm and 14 mm) to suppress Bragg peaks from the sample environment such as from cryostats, furnaces, magnets or high pressure cells (< 14 kbar) and (< 100 kbar).

For the fine sample positioning in the scattering plane, there is a motorized xy-table controlled by computer. The accuracy of the sample positioning with respect to the detector center of about 0.5 mm is achieved by a special measurement of the standard sample and quick automatic refinement by a script. The positioning is very important for accurate determination of atomic displacement parameters ADPs.

HRPT is also equipped with computer controlled sample changers for either eight samples at room temperature or for four samples for the temperature range of (1.5–315) K.

A very small leak in the detector results in a slow continuous decrease in the gas mixture pressure and worsening of the detector PHS

spectrum. Therefore a special cleaning/ pressurizing system has been designed at PSI and manufactured by the MESSER Schweiz company. The system allows for effective cleaning of the gas mixture and removes the impurites such as O_2, N_2, H_2O, etc. by a circulation of the gas mixture through appropriate filters without a need for pumping out the detector.

The instrument is controlled via the SICS and SEA softwares developed by LDM/PSI for a UNIX workstation, permitting fully automatic computer controlled measurements, data reduction and rapid online refinements.

The HRPT instrument is not only used for academic science in the frame of the SINQ user policy program, but also a limited amount of HRPT beam time is available to interested industrial companies. Certain companies cannot disclose the details of their research for confidentiality reasons, and in this case HRPT beam time can be purchased according to the PSI rules.

A further improvement of HRPT would be a second monochromator such as Ge, optimized for 2.45 Å.

6. SINGLE-CRYSTAL NEUTRON DIFFRACTOMETER TRICS AT SINQ [b d g]

The single crystal neutron diffractometer TriCS [9] (Figs. 23, 24), see also http://www.psi.ch/sinq/trics/, has been designed for solving structural problems in chemistry (λ=1.18 Å, Ge(311), maximum $\sin(\theta/\lambda) = 0.7$ Å^{-1}) as well as in magnetism (λ=2.31Å, PG(002)). It has been successfully operated for 15 years on a thermal beam tube at SINQ.

Unique features are the tilting option allowing bulky equipment such as magnets and the possibility to switch within minutes from a single tube ^{3}He detector to a two-dimensional area detector (160 mm by 160 mm, radial collimator, time-delay readout).

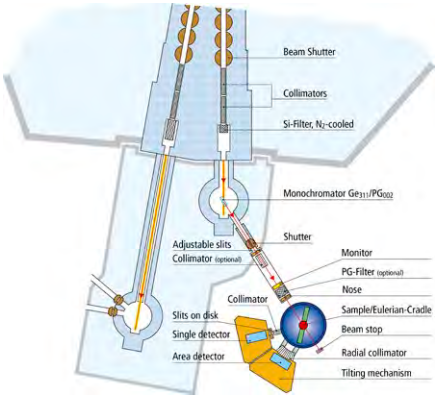


Figure 23: Present layout of the single crystal neutron diffractometer TriCS@SINQ.

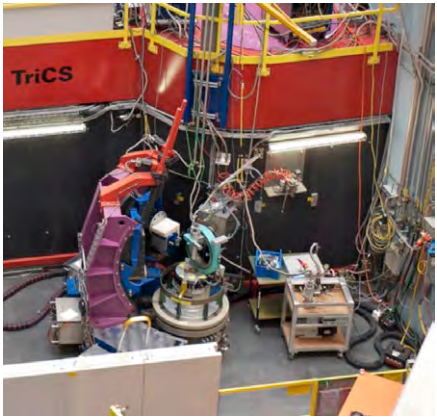


Figure 24: Present layout of the single crystal neutron diffractometer TriCS@SINQ.

Future developments in progress will not only increase the flux by an improved primary optics and the new vertically focusing PG monochromator (SwissNeutronics), to be installed in 2013, but also dramatically reduce the background as a result of improved shielding, based on state-of-the-art absorption calculations. A key issue in this new instrument ZEBRA (Fig. 25) – presently in the predesign phase – will be the optional analyser in front of the single detector. ZEBRA will also yield much faster data collection by removing air cushions. The complete unmagnetic construction will allow higher magnetic fields up to 12 Tesla. ZEBRA also will be suitable for smaller crystal volumes as required by investigations of novel materials, for example in the field of multiferroics.

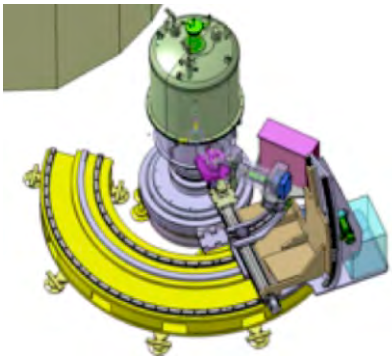


Figure 25: ZEBRA, the new single crystal neutron diffractometer at SINQ (design phase), replacing TriCS.

New software developments will assist less experienced users to benefit from all the options available. We continue here improvements such as possibilities to create 3D-cuts in q-space for TriCS data collected with the 2D-detector.

In summary, the new ZEBRA will focus on investigation of magnetic structures with the possibility to use externel magnetic fields up to 12 Tesla, but also will improve crystallographic investigations on dedicated systems as presently covered by TriCS, with lower data collection times.

7. HEIMDAL HYBRID NEUTRON SPECTROMETER PROJECT AT THE EUROPEAN SPALLATION NEUTRON SOURCE ESS: PROBING MULTIPLE LENGTH SCALES IN ONE INSTRUMENT [g i k b j]

Ongoing improvements in material performances are reached for example by the incorporation of advanced ceramics and polymers into heterogeneous systems. Their performances usually depend on the interplay between properties defined by the atomic, nano/mesoscopic and microscopic structure. Traditionally such structural information is collected in separated experiments such as wide angle diffraction (probing the atomic scale, $0.3\ Å^{-1} \leq Q \leq 50\ Å^{-1}$), small angle scattering (nano/meso scale, $0.002\ Å^{-1} \leq Q \leq 0.1\ Å^{-1}$) and direct space imaging techniques (submillimeter to millimeter scale).

The hybrid instrument HEIMDAL [10] (Fig. 26), is proposed by a collaboration of the universities of Aarhus and Copenhagen as well as the LNS, to be built at the European Spallation Neutron Source ESS (Lund, Sweden).

The instrument is designed to obtain a coherent multi-length scale picture of these materials. The idea is to merge neutron pow-

der diffraction (probed length ζ ~ 0.01–5 nm), small angle neutron scattering (ζ~1–500 nm) and neutron imaging (ζ~0.01–100 mm), giving a huge advantage, especially for in situ-measurements. To fit these needs, the instrument will have two guide systems looking on different parts of the source (thermal and cold) through a single beam port.

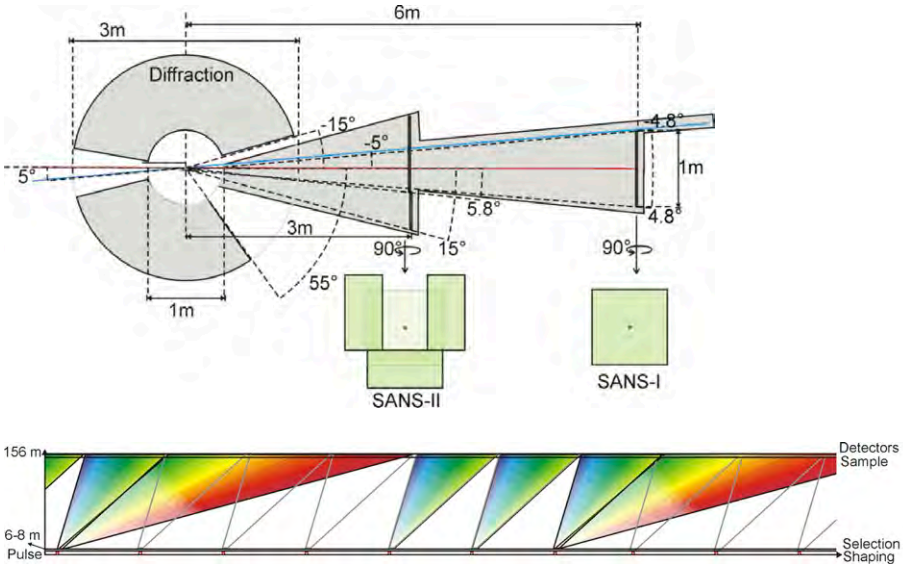


Figure 26: A schematic illustration of the combined powder diffraction and SANS setup. Below is the pulse train, where three diffraction pulses are skipped to allow a longer SANS pulse. Other operations modes are possible depending on the used choppers sequence. The short wavelength pulse and the long wavelength pass are transported through different guides due to different needs for the neutron optics.

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