Neutron Powder Diffraction with Long Pulses at the European Spallation Source

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European Spallation Source
Neutrons are special

**Charge neutral**
Deeply penetrating

**S=1/2 spin**
probe directly magnetism

**Nuclear scattering**
Sensitive to light elements and isotopes

Li motion in fuel cells

Solve the puzzle of High-Tc superconductivity

Actives sites in proteins

Help build electric cars

Efficient high speed trains

Better drugs
Major National and International Neutron Sources

- Operating Spallation Sources
  - LANSCE
  - SNS (2006)
  - SNS-TS2

- Operating Reactor Sources
  - NCNR
  - HIFAR

- Planned Spallation Sources
  - FRM-II
  - SINQ
  - J-PARC (2007)
  - OPAL (2007)

- Planned Reactor Sources
  - ILL
  - Dubna
  - CNS
  - CSNS
  - Bhabha

- Other Sources
  - ISIS TS1&2
  - ESS
  - LANSCE
  - NCNR
  - HIFAR
  - SNS (2006)
  - SNS-TS2
  - J-PARC (2007)
  - OPAL (2007)
  - Brazilian Neutron Source
  - Argentinian Neutron Source
First neutron instruments

1950s
The Next Generation at ILL

2D detectors for CW Powder Diffraction

UK-EPSRC Super-D2B project at ILL

- 128 Detectors
- Relative long detectors
- Need to integrate over Debye Scherer Rings

E. Suard, C. Ritter, A. Hewat, P. Attfield... (Edinburgh)
Alan Hewat, Super-D2B, EPDIC-IX, Prague, 3 Sept 2004
D20 at ILL

- Large monochromator viewing a large beam cross section
- Various $\lambda$ available
- Large continuous coverage micro-strip detector
  - Major technological challenge
- Relative ease in changing take-off angle from low to high resolution.
- Flexible set-up in special environments
  - Pressure cells
  - Reaction cells
  - Furnaces...
Flexible resolution on D20

\[ \text{Na}_2\text{Ca}_3\text{Al}_2\text{F}_{14} \text{ in q-space } (4\pi\sin\theta/\lambda) \]

60s Cu(200) 1.297 Å at 42° takeoff (black)
120s Ge(117) 1.36 Å at 120° takeoff (red)

Overlap at high Q makes structural refinements impossible with high-flux data
Fast data collection / processes

Ti$_3$SiC$_2$ made by hot isostatic pressing is expensive

In-situ investigation of thermal explosion synthesis (TES)

Initiate by heating from 850-1050 °C at 100 °C/min

Acquisition time 500 ms (300 ms deadtime)

Experiment band-structures showing that the Fermi-surface nesting destabilizes with pressure (like with doping x) to allow superconductivity to emerge.
Two ways to produce Intense Neutron Beams Safely

**Fission:** One neutron in, three neutrons out

**How:** Use a nuclear reactors

**Spallation:** Up to 30 neutrons per proton!

**How:** Use an accelerator to propel a proton onto a tungsten element target
Anatomy of a Pulsed Spallation Neutron Source

Ion Source → Accelerator → Compressor/Accumulator → Target

- Current: ≈ 20 mA
- Time: ≈ 1 ms
- Current: ≈ 20 A
- Time: ≈ 1 μs

Pulsed Neutron Beam (TOF)
Moderators

High Energy Neutrons

Proton Pulse

FWHM ~ 1µs

time
Moderators

Beam Port

Target

Proton Pulse

FWHM ~ 1µs

Δt (µs)

λ (Å)

Δ (µs)

1 Å

2 Å

5 Å

0

100

200

300

time (µs)

0

5.0

10.0

15.0

20.0

2.0

3.0

0

1

2

3


time

APD-2013 | NIST | Dimitri Argyriou
POLARIS + HRPD (ISIS)

POLARIS 1991

POLARIS 1996-2010
terns recorded hold temperature study," has been confirmed anisotropy included rate constant shown K, can be seen of aging OL of 11.

Table a, is shown in Table of c-phase, t-phase, the increase shown aging in 1100°C; the amount of c-phase, t-phase, OL of 11.

The shown increase in amount of aging phase amounts fits exponential functions, the c-phase, t-phase, the aging corresponds to the c-phase, t-phase, the values in Table a, the value of Lattice parameter OL of 11.

Figure 3. Lattice parameter versus time (hours) for different stabilizers content line. The peak broadening at c-ZrO, did not change, the peak broadening at t-ZrO, 0.6% during aging as expected given the lenticular shape.

Analyses by the Rietveld method provide rather precise lattice parameter values.

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Step change in instrumentation

- High count rate / efficiency
- Large detector arrays
- Optimised beam transport
- Flexible resolution

late 1990s
Understanding the Insulating Phase in Colossal Magnetoresistance Manganites: Shortening of the Jahn-Teller Long-Bond across the Phase Diagram of La$_{1-x}$Ca$_x$MnO$_3$


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- High throughput and high quality data allows for detailed parametric investigation as a function of composition, temperature, pressure etc

- Was tour de force, now routine!
1995 cf. 2013

- 1995 500 mg 24+ hrs
- 2013 500 mg 15-20 minutes with increased Q-range
Polycrystalline Diffractometers
Resolution & Intensity

Diffractometers in the World

Several instruments to be constructed at once
Relation to other facility is important
Requirements in Modern NPD

• Flexible Instrumentation
  - trade flux for resolution
  - Match resolution to problem
  - Match Q-range to system of interest

• Smaller samples
  - Isotopic substitution
  - Extreme conditions (high pressure, high magnetic fields, simultaneous measurements)

• Fast data collection
  - Parametric studies of phase diagrams
  - In situ study of reactions
  - Kinetic studies

• New technical developments
  - Polarisation
  - Hydrogenous materials

Reactor and Spallation Sources have distinct advantages and disadvantages

- Take-off Angle
- Moderator pulse shape

Need for higher source fluxes, how?

Advanced data acquisition electronics and methods to track fast reactions.

- micro-secs possible from the perspective of flux
- Problems in using large detector arrays in this way

Polarisation is flux intensive!
Instruments
22 Instruments in construction budget

Proton Accelerator
Energy: 2.5 GeV
Frequency: 14 Hz
Current: 50 mA

Target Station
Solid Rotating W
He or Water Cooled
5MW average power
>22 beam ports

5 times more powerful than SNS
30 times brighter than ILL

Total Cost of Project
1843 (2013) Mil €
An International Collaboration

Sweden, Denmark and Norway: 50% of construction and 20% of operations costs

European partners pays the rest
ESS
- "operations phase"

ESS AB
- "construction phase"

ESS AB
- "design update phase"

ESSS Secretariat
- "campaign phase"

2007

2010

2013

2014

2019

2025-2030

• Deliver Technical Design Review

• Start Civil construction

• Deliver Conceptual Design Review

• Costing Report

• Cost Book

• Win international support for building ESS in Lund, Sweden

ESS reaches full operational capability at 5MW and 22 instruments
Proton Beam

ESS reference suite of 22-Instruments
Advantages of cylindrical shape:
- Better for serving many instruments – wide angle beam extraction
- Highest neutronic performance

ESS Moderator

Cold Moderator

Thermal Moderator
ESS Long Pulse Structure Compared to Other Sources

Figure 2.19: Single-pulse source brightness as a function of time at a wavelength of 5 Å at ESS, ILL, SNS, J-PARC and ISIS target stations 1 and 2. In each case, the cold moderator with the highest peak brightness is shown. The peak brightness at ESS will be higher than that of any of the short pulse sources, and will be more than an order of magnitude higher than that of the world's leading continuous source. The time-integrated brightness at ESS will also be one to two orders of magnitude larger than is the case at today's leading pulsed sources.

The designs of instruments at ESS will be less limited by the time-structure of their long-pulse source than are instruments at short-pulse sources. They will benefit from a substantially higher peak brightness, combined with a time-average brightness which is much higher than that at any short-pulse source, while retaining much of the flexibility of continuous-source instruments.

Many of the instruments will be substantially longer than their counterparts at short-pulse sources. The underlying reason is the requirement for good wavelength resolution; the pulse length at the source represents the uncertainty in the emission time of the neutrons, which can be reduced compared to their time-of-flight by making the instrument longer. However, the instrument length also directly affects the bandwidth; that is, the longest wavelength that can be measured from a particular source pulse before it overlaps in time with the shortest wavelength emitted from the following pulse. Pulse-shaping choppers are an alternative method of improving wavelength resolution. Placed close to the source, they effectively reduce the pulse length, and hence the resolution, without making it necessary to increase instrument length. A large part of the optimisation of the instrument suite consists of balancing resolution and bandwidth considerations through appropriate combinations of instrument lengths and chopper systems.

2.3.1 White-beam instruments

The majority of the instruments at ESS will use a substantial part of the full white beam. Their bandwidth will be limited by their length and choice of pulse suppression, and it will be possible to tailor their resolution using a pulse-shaping chopper. They fall into two categories – large pulse width and small pulse width – depending on the required pulse width compared to the intrinsic length, $\tau = 2.86$ ms, of the neutron pulse.

Large pulse width = $\tau$ SANS, spin-echo, macromolecular crystallography, and particle physics: There are seven instruments in this category in the ESS reference suite, all of which are well suited to the long-pulse time structure. They can use the full ESS pulse width and thus benefit from the high peak and time-average brightness. The ESS instruments will significantly outperform equivalent present-day instruments, due to the often unnecessarily good wavelength resolution of those instruments at contemporary short-pulse sources, and to the inherently lower peak brightness available to instruments at continuous sources.
Two Strategies for Neutron Instrumentation at ESS

Use as much as possible of the whole pulse:
Good for low wavelength resolution instruments.
SANS, Reflectometry, single crystal diffraction.
Estimated gains 10-100 times than currently available.

Cut the long pulse into smaller pulses:
Good for higher wavelength resolution instruments
Diffraction, cold/thermal spectrometers.
Long Instruments (80-100 m)
Estimated gains 10-30 times than currently available.
Thermal gains lower.
How to do it?

- The long pulse is too broad to use for diffraction studies.
  - Choppers will play the role of the moderator-response time in a conventional short pulse source.
  - Important to get the first pulse shaping chopper as close as possible to moderator.
  - Tunable wavelength range (1.9 Å band up to a maximum of 6 Å)
  - Can ‘slew’ choppers to cover complete wavelength band in several pulses
  - Tunable Δλ/λ (from < 0.02% - 5% at λ = 1.45 Å) with PSC
  - Resolution/peak shape not determined by moderator time constant
Pulse Shaping Chopper

Intercity

0 1 2 3
time (ms)

ILL
Instrument Workshop Summary

• Recommended Phase I Diffractometers
  • Single-crystal diffractometer for macromolecular crystallography
  • Single-crystal diffractometer for magnetism
  • Narrow-bandwidth, high-resolution tunable powder diffractometer

• Suggested for further consideration
  • Hybrid diffractometer
  • Structured pulse engineering spectrometer
  • Single-crystal (and/or powder) diffractometer for extreme conditions

Debated at the Experts’ Meeting at Vaals (September) and SAC (November 2010)
Concepts for powder diffractometers at ESS

Use source peak brightness ➔ TOF wavelength band instrument (conventional spallation source instrument)

Use source time-average brightness ➔ Monochromator instrument with TOF detector (reactor-like instrument with enhanced capabilities)
Thermal powder TOF diffractometer

- Up to 1.9 Å single frame wavelength band (normal mode 0.5 – 2.4 Å)
- High flexibility cf. SPSS instrument
  - Tuneable wavelength range (1.9 Å band up to a maximum of 6 Å)
  - Can ‘slew’ choppers to cover complete wavelength band in several pulses
  - Tuneable $\Delta \lambda / \lambda$ (from < 0.02% - 5% at $\lambda = 1.45$ Å) with PSC
  - Tuneable flux with PSC
  - Resolution/peak shape not determined by moderator time constant
- Long instrument (156 m)
  - Low background
Bispectral Powder Diffractometer

- Bispectral extraction (0.8 – 10 Å)
- Wavelength frame multiplication gives 3.8 Å wavelength band
  - Normal mode (0.8 – 4.6 Å)
  - Tuneable wavelength range
- Beat chopper
  - Flexible resolution
  - Flexible flux
- Shorter instrument (75m cf. 150m)
- Complementary with thermal powder diffractometer
Pulsed monochromatic powder diffractometer

- Like a reactor instrument in appearance
  - $Q_{\text{max}} \sim 12.5 \text{ Å}^{-1}$
  - Variable resolution (takeoff angle)
  - Trade flux for resolution (takeoff angle)
  - But longer instrument (total flight path 50 m cf. 22m at D20)
  - And optimised beam transport

- Background suppression
  - Only integrate around elastic line

- New capabilities cf. reactor instrument
  - Multi-wavelength data collection
  - Separate coherent/incoherent scattering
  - Elastic/inelastic measurements
  - Fast kinetics
Neutron Diffraction and ESS

Tuning of the resolution to the requirements of each experiment by adjusting the opening time of the pulse-shaping chopper. This unique capability is illustrated in Figure 2.24 where the resolution and intensity of a range of neutron powder diffractometers at leading large-scale facilities around the world are shown. The performance of current instruments is indicated by various symbols, showing their specialisation for high flux or high resolution, while the ESS diffractometers trace out continuous lines along which the instrument performance can be selected, depending on the needs of the experiment. The ESS instruments match the projected best performance of today's new instruments, though there are limitations in comparison to short-pulse sources at large \( Q \) (\( 20^\circ A^1 \)), which do not appear in such a figure. At ESS, the thermal powder diffractometer covers the core crystallography and in situ processing science case for thermal neutrons up to a \( Q_{\text{max}} \) of around 20 \( A^1 \). The pulse-shaping chopper allows the wavelength resolution to be matched to the experimental requirements over a very wide range, in the best case achieving a resolution of <0.01%. The bispectral powder diffractometer can access longer wavelengths and is well-suited for powder magnetic structure determination, larger unit cell powder crystallography and in situ experiments in the intermediate \( Q \) range (\( Q_{\text{max}} < 13^\circ A^1 \)). The pulsed monochromatic powder diffractometer is a high-throughput instrument for in situ chemical processing, crystallography, pole figures, high time-resolution kinetics and hydrogen-containing materials, with the option for variable resolution inelastic scattering mapping. It uses a set of crystal monochromators with access to a wide range of takeo angles to tune the accessible \( Q \) range, flux and instrument resolution. The materials science & engineering diffractometer will allow users to study mechanical and micro-structural properties, texture, phase transformations and kinetics in engineering and functional/smart materials. Dedicated sample environment will allow a wide range of conditions to be probed. Structural studies include the determination of residual stresses in engineering materials and components, an important area in welding and joining R&D. The extreme conditions instrument is built around bulky, complex sample environment (high pressure, high magnetic field) and the limitations that imposes on detector coverage and geometry. It is a multi-role instrument to perform diffraction, spectroscopy, SANS and imaging experiments. For all the diffractometers, the resolution and bandwidth can be tailored to each application.
Diffraction at ESS

• ESS will offer levels of versatility not available in any type of diffractometer either at reactor or short-pulse spallation sources.
• Mechanical chopper systems provide for the
  • Wavelength resolution
  • Wavelength band
  • Peak shape
• ESS diffractometers will require significant software developments for data reduction and analysis
• Emphasis in instruments will be towards sample environments, following current trends
• To understand results scientific computing is becoming increasingly important