

# THE PROSPECT OF NEUTRON SCATTERING IN THE 21<sup>ST</sup> CENTURY: A POWERFUL TOOL FOR MATERIALS RESEARCH

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## ABSTRACT

**THE PROSPECT OF NEUTRON SCATTERING IN THE 21<sup>ST</sup> CENTURY: A POWERFUL TOOL FOR MATERIALS RESEARCH.** Over the last 60 years research reactors (RRs) have played an important role in technological and socio-economical development of mankind, such as radioisotope production for medicine, industry, research and education. Neutron scattering has been widely used for research and development in materials science. The prospect of neutron scattering as a powerful tool for materials research is increasing in the 21<sup>st</sup> century. This can be seen from the investment of several new neutron sources all over the world such as the Spallation Neutron Source (SNS) in USA, the Japan Proton Accelerator Complex (JPARC) in Japan, the new OPAL Reactor in Australia, and some upgrading to the existing sources at ISIS, Rutherford Appleton Laboratory, UK; Institute of Laue Langevin (ILL) in Grenoble, France and Berlin Reactor, Germany. Developing countries with moderate flux research reactor have also been involved in this technique, such as India, Malaysia and Indonesia. The Siwabessy Multipurpose Reactor in Serpong, Indonesia that also produces thermal neutron has contributed to the research and development in the Asia Pacific Region. However, the international joint research among those countries plays an important role on optimizing the results.

**Keywords:** Spallation neutron source, Research reactor, Neutron scattering

## INTRODUCTION

Soon after, the discovery of neutron by James Chadwick in 1932 [1], two research groups in Europe demonstrated the technique of neutron diffraction [2]. But studying materials using the diffraction technique only became feasible years later as high-flux thermal neutron sources produced from reactors became available. Clifford Shull and Bertram Brockhouse, who received the 1994 Nobel Prize in Physics performed their pioneering contributions to the development of neutron scattering techniques for studies of condensed matter. They developed the neutron diffraction and neutron spectroscopy techniques, respectively in the 1940s and 1950s, which in simple terms, they helped answer the questions of where atoms "are" and of what atoms "do" [3,4]. Neutron scattering techniques have played an important role on investigating the microscopic origins of materials' physical, electrical, magnetic, chemical, and biological properties. Unlike beams of x rays and electrons, which interact strongly with a material's electrons, the neutron's fundamental attributes make it a unique and complementary probe.

The neutron can be produced either from the research reactor or the spallation neutron sources. The earliest neutron sources were based on radioactive decay. Their successors, the nuclear fission reactors, produced fluxes whose peak values quickly reached a plateau in the 1950s. Advances in instrumental energy resolution and detector efficiencies continue to make neutron reactors useful. The Spallation Neutron Source is the culmination of a line of proton-driven pulsed neutron sources, whose neutron fluxes and efficiencies now exceed continuous sources. The demand of using neutron as probe has been increasing for the last decades, due to very wide application in material science ranging from polymer, biology, metallurgy, crystallography etc. The investment of new neutron sources in several developed countries, such as Spallation Neutron Source (SNS) in USA, Japan Proton Accelerator Research Complex –Tokaimura Japan, and the new OPAL Reactor in Australia, have become good sign for the scientific opportunity in the near future. This article is a brief review about the neutron scattering, started by a basic idea why neutron is important probe. It is then followed by the source of neutron and the historical development of neutron sources all over the world. The basic principles of neutron experiments and the scientific opportunity of using neutron will also be explained, followed by several examples of research performed by neutron scattering.

## WHAT IS A NEUTRON?

The neutron is a powerful tool for the study of condensed matter (solids and liquids) in the world around us, having significant advantages over other forms of radiation in the study of microscopic structure and dynamics. Neutron scattering gives detailed information about the microscopic behavior of condensed matter, playing a major role in shaping the experimental and theoretical understanding of materials ranging from magnetism and superconductivity to chemical surfaces and interfaces.

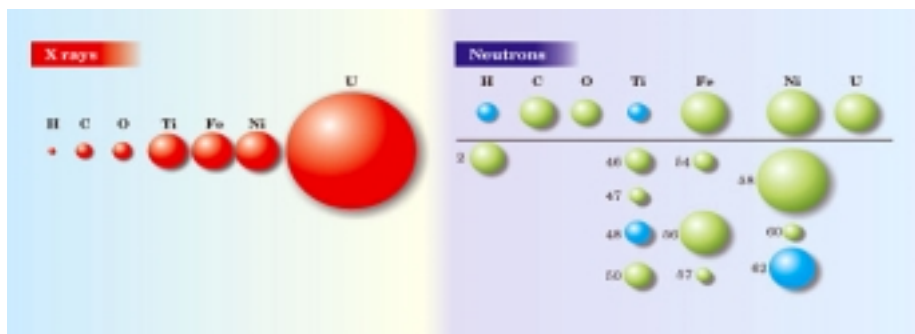


Figure 1. Comparison of x-rays and Neutrons.

A neutron has no charge, has an electric dipole moment that is either zero or extremely close to it, and interacts with atomic nuclei through the very short-range nuclear force [5]. Consequently, a neutron beam penetrates matter much more deeply than an x-ray or electron beam can. Indeed, many neutrons in the beam will pass completely through the material. However, the ones that interact directly with atomic nuclei or with any unpaired electron spins in the material get deflected from their original path [6].

Another desirable property of neutrons is their unique sensitivity to hydrogen atoms. The pronounced scattering cross section means that neutrons can be used to precisely locate hydrogen atoms and provide an accurate determination of a compound's molecular structure, information that is important for the design of new therapeutic drugs, biomolecule, and also polymers and other types of hydrogenous materials [7]. Neutrons can locate other light atoms among heavy atoms as well. That capability was essential in determining the chain ordering of oxygen atoms in high-temperature superconductors and of other complex oxides and minerals. X rays scattered from such oxide compounds, in contrast, are much less sensitive to the oxygen atoms amidst their heavy neighbors.

Figure 1 shows a comparison of x-ray and neutron scattering cross sections for an identical set of elements and their more common isotopes. The size of each circle is a measure of the relative cross section, and mass numbers identify the different isotopes, with the top row representing an isotopically average value. The systematic variation in the x-ray scattering cross sections occurs because x rays scatter from electrons, the number of which increases monotonically across the periodic table. Neutrons scatter from nuclei. Thus, the cross section varies in a way that depends on the nuclear structure.

Some isotopes, including the ones colored blue here, exhibit negative scattering length. The absence of a systematic variation in neutron cross section across the periodic table is a testament to the complementary information that neutrons provide [6,7].

## NEUTRON SOURCES

Neutrons for scattering experiments can be produced either by nuclear fission in a reactor or by spallation when high-energy protons strike a heavy metal target (W, Ta, or U) (see Figure 2). In general, reactors produce continuous neutron beams and spallation sources produce beams that are pulsed between 20 Hz and 60 Hz. The energy spectra of neutrons produced by reactors and spallation sources are different, with spallation sources producing more high-energy neutrons. About 1.5 useful neutrons are produced by each fission event in a Nuclear Reactor, whereas about 25 neutrons are produced by spallation for each 1-GeV proton incident on a Tungsten target.

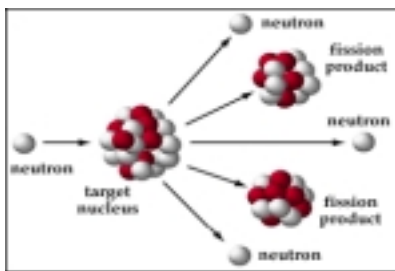


Figure 2. Nuclear Fission.

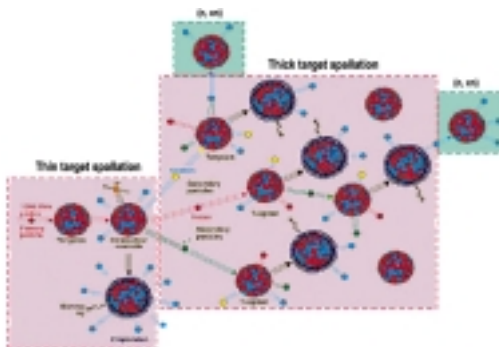


Figure 3. Spallation Process.

## Spallation

A heavy metal target is bombarded with pulses of highly energetic protons from a powerful accelerator, driving neutrons from the nuclei of the target atoms. This results in an extremely intense neutron pulse, delivered with only modest heat production in the neutron target. The spallation target is made from the heavy metal tantalum. Protons hitting nuclei in the target material trigger an intranuclear cascade, placing individual nuclei into a highly excited state. The nuclei then release energy by evaporating nucleons (mainly neutrons), some of which will leave the target, while others go on to trigger further reactions. Each high energy proton delivered to the target results in the production of approximately 25 neutrons [8].

Neutrons from reactors and spallation sources must be moderated before being used for scattering experiments. The energies and wavelengths of the neutrons produced from reactors or spallation sources must first match the energies of elementary excitations and length scales of interatomic distances in materials of interest. Neutron-scattering facilities throughout the world produce neutrons with energies of tens or hundreds of MeV. That's far too high for investigating condensed matter. To create a beam of "thermal" neutrons, one must first cool the neutron beam typically to room temperature, around 25 meV by passing it through a water bath known as a moderator. When the beam's wavelength matches the interatomic spacing of typical materials, it becomes possible to probe excitations across a range of length scales and achieve good energy resolution [6].

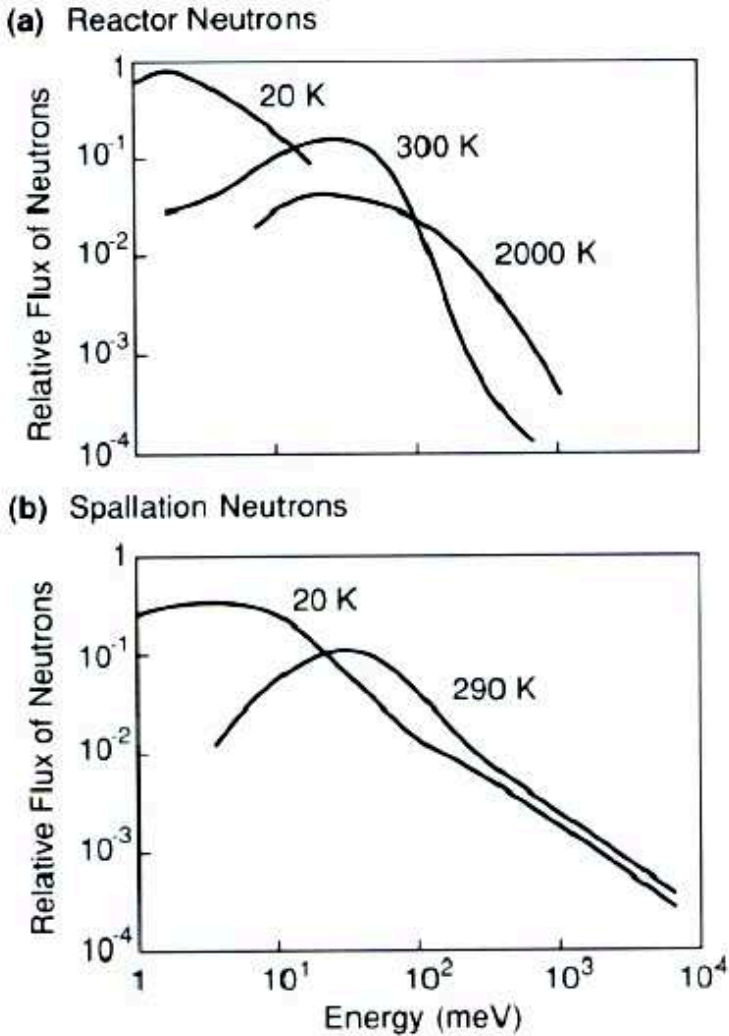


Figure 4. Neutron energy spectra.

Figure 4 shows the neutron spectra produced from reactor and spallation. The reactor spectra are Maxwellian while the Intensity and peak-width  $\sim 1/(E)^{1/2}$  at high neutron energies at spallation sources. Cold sources are usually liquid hydrogen (though deuterium is also used at reactors and methane is sometimes used at spallation sources). Hot source at ILL (only one in the world) is graphite, radiation heated.

Table 1. A Comparison of Reactors and Spallation Sources.

Short Pulse Spallation	Source Reactor
Energy deposited per useful neutron is ~20 MeV	Energy deposited per useful neutron is ~ 180 MeV
Neutron spectrum is “slowing down” spectrum – preserves short pulses	Neutron spectrum is Maxwellian
Constant, small dl/l at large neutron energy => excellent resolution especially at large Q and E	Resolution can be more easily tailored to experimental requirements, except for hot neutrons where monochromator crystals and choppers are less effective
Copious “hot” neutrons=> very good for measurements at large Q and E	Large flux of cold neutrons => very good for measuring large objects and slow dynamics
Low background between pulses => good signal to noise	Pulse rate for TOF can be optimized independently for different spectrometers
Single pulse experiments possible	Neutron polarization easier

## HISTORY DEVELOPMENT OF NEUTRON SOURCES

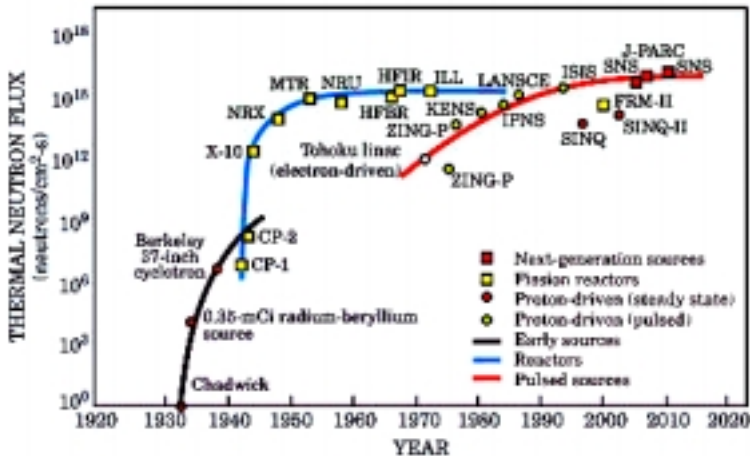


Figure 5. Historical Development of Neutron Sources.

The limitation on thermal-neutron flux from the core of the reactor during fission, has been made the researchers to find another source. In the 1950s the researchers consider using a high-flux spallation source to produce nuclear materials. In 1980 the first spallation neutron source, a Neutron Science Laboratory (KENS) at Japan's High Energy Accelerator Research Organization (KEK) was constructed and the Intense Pulsed Neutron Source

(IPNS) at Argonne National Laboratory (ANL) a year later. By the late 1970s, Europeans were increasingly dominating the neutron-scattering community, with developments at the Institut Laue-Langevin (ILL) in Grenoble, France, and a network of national facilities [11]. The US installed a cold source, "guide hall," and associated instruments at what is now the NIST Center for Neutron Research in Maryland. [12]. The continuous Swiss Spallation Neutron Source (SINQ) facility was also constructed at the Paul Scherrer Institut in Switzerland. However, worldwide, accelerator based spallation sources of neutrons are much less numerous than reactor based ones.

The SNS at Oak Ridge [13,14] and the spallation target currently under construction as part of the Japan Proton Accelerator Research Complex (J-PARC) facility in Tokai represent the next generation of spallation sources [15]. Although efforts in Europe to build a next-generation spallation source have not yet led to a funded project, [10] a significant upgrade is under way at ISIS. In 2005, China granted approval for work to begin on the Beijing SNS, a facility sponsored by the Chinese Academy of Sciences.

Meanwhile, the Australia has also constructed new OPAL research reactor at ANSTO. Both reactors and spallation sources are expensive to build and require sophisticated operation. SNS at ORNL will cost about \$1.5B to construct and ~\$140M per year to operate. Either type of source can provide neutrons for 30-50 neutron spectrometers. This experiment shows how the small science needs a large facilities [6,13]. Figure 5 shows the historical development of thermal-neutron sources, both continuous and pulsed. Figure 6 shows the neutron facility at ILL, Grenoble, France including the European Synchrotron Radiation Facility (ESRF).



Figure 6. Research Reactor, ILL and European Synchrotron Radiation Facility, Grenoble, France.

# WHAT DO WE NEED TO DO A BASIC NEUTRON SCATTERING EXPERIMENT?

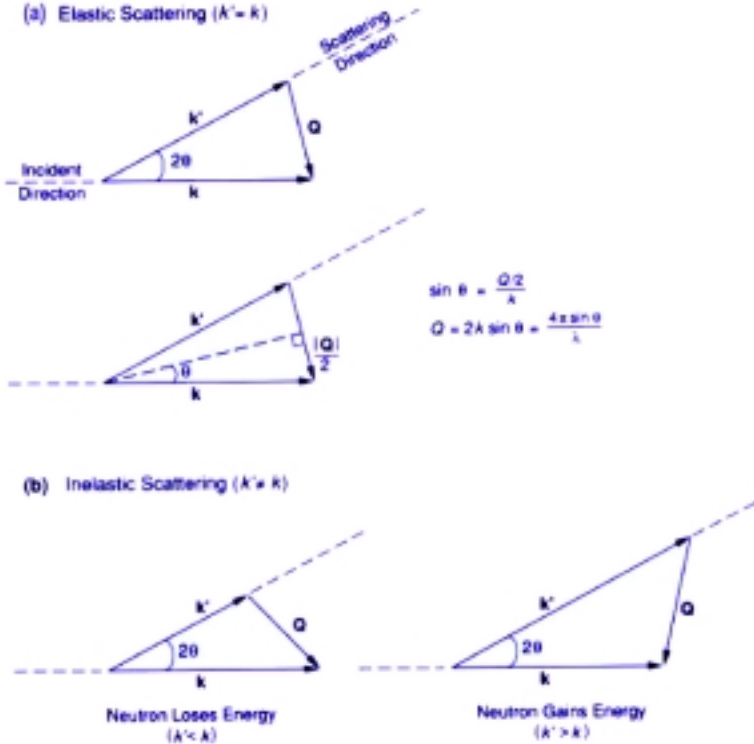


Figure 7. Basic neutron experiment.

In order to perform neutron scattering experiment, we need a source of neutrons, a method to prescribe wave vector of neutrons incident on the sample, and a method to determine the wavevector of the scattered neutron, for the inelastic case and a neutron detector. Remember: wavevector,  $k$ , and wavelength,  $\lambda$ , are related by:

$$k = mnv / (h/2\pi) = 2\pi / \lambda \tag{1}$$

In general, neutron scattering experiments measure the number of neutrons scattered by a sample as a function of the wave vector change ( $Q$ ) and the energy change ( $E$ ) of the neutron. Expressions for the scattered neutron intensity involve the positions and motions of atomic nuclei or unpaired electron spins in the scattering sample. The scattered neutron intensity as a function of  $Q$  and  $E$  is proportional to the space and time Fourier Transform of the probability of finding two atoms separated by a



particular distance at a particular time. Both function of Q and E followed the conservation of momentum and energy, as follows :

$$Q = k' - k \tag{2}$$

$$E = ( h^2 m / 8 \pi^2 ) (k'^2 - k^2) \tag{3}$$

And the scattering properties of sample depend only on Q and E, not on neutron.

The elastic scattering is measured the correlation of atomic position such as the static structure factor in the liquid and follows the relation :

$$S(Q) = 1 + \int g(r) e^{-iQ \cdot r} dr \tag{4}$$

Neutrons can also gain or lose energy in the scattering process: this is called inelastic scattering. The inelastic scattering follows the relation :

$$S(Q, \omega) = 1/h \iint G(r,t) e^{i(Q \cdot r - \omega t)} dr dt \tag{5}$$

### THE 1994 NOBEL PRIZE IN PHYSICS

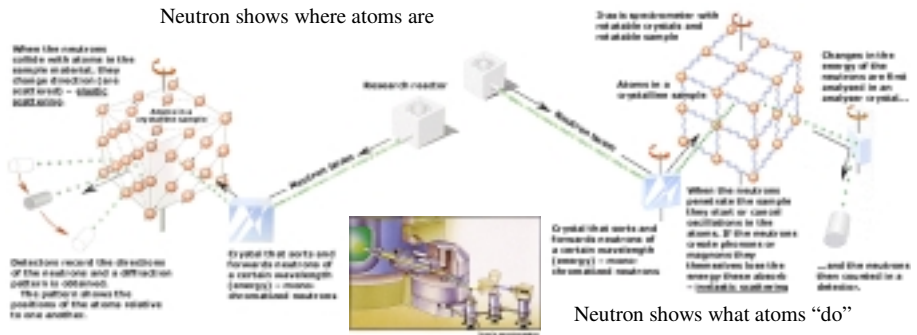


Figure 8. 1994 Nobel Prize to Schull and Brockhouse.

Based on the two principles in the neutron scattering, Clifford Shull and Bertram Brockhouse, performed their pioneering contributions to the development of neutron scattering techniques for studies of condensed matter. They developed the neutron diffraction and neutron spectroscopy techniques, respectively in the 1940s and 1950s, and were awarded the 1994 Nobel Prize in Physics . In simple terms, they helped answer the questions of where atoms "are" and of what atoms "do". (see <http://www.nobel.se/physics/educational/poster/1994/neutrons.html>).

## Scientific Opportunity by Neutrons

The various neutron-scattering instruments installed on the worldwide neutron facilities will determine the range of topics that both reactor and spallation neutron sources address. Each instrument is narrowly optimized to meet the measurement objectives a particular range and resolution of energies and momenta transferred from neutrons to the sample of interest to suit a specific class of scientific problems. In general, there are two typical of neutron instruments, neutron diffraction and spectrometer.

**Diffraction.** Diffractometers look at structures, that is the spatial distribution of atoms, molecules, or larger scale structures. They measure neutrons that have been scattered elastically from a sample, ie. energy has not been exchanged between the neutron and the sample. This process is known as diffraction.

**Spectroscopy.** Spectrometers measure inelastic scattering, ie the change in energy of a neutron when it scatters in from sample, and relate-this to the atomic dynamics of the sample. This process is known as spectroscopy.

Many types of neutron scattering spectrometer are required because the accessible Q and E depend on neutron energy and because resolution and detector coverage have to be tailored to the science for such a signal-limited technique. There are also examples of Specialization of Spectrometers: Optimizing the Signal for the Science.

- **Small angle scattering**

Small diffraction angles to observe large objects ~ long (20 m) instrument.

- **Back scattering**

instrument with a very good energy resolution ( $\sim$ neV) ~ perfect crystal analyzer.

Time-of-flight Methods Can Give Complete Dispersion Curves at a Single Instrument Setting in Favorable Circumstances, i.e. CuGeO<sub>3</sub> is a 1-d magnet as shown in Figure 11. With the unique axis parallel to the incident neutron beam, the complete magnon dispersion can be obtained.

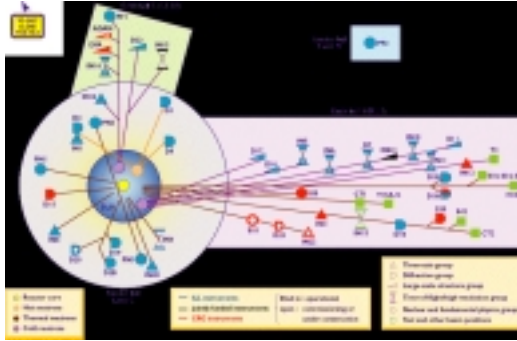


Figure 9. Neutron Instruments at the experimental and guide hall reactor, ILL.



Figure 10. A ~ 30 X 20 m2 Hall at the ILL Houses About 30 Spectrometers.

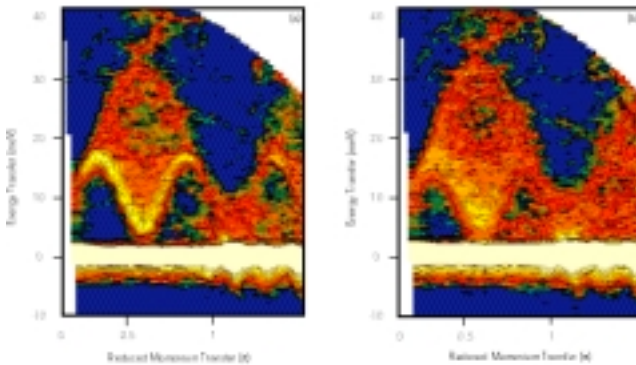


Figure 11. The complete magnon dispersion for CuGeO<sub>3</sub> is a 1-d magnet measured by using a time-of-flight neutron scattering experiment at the spallation source.

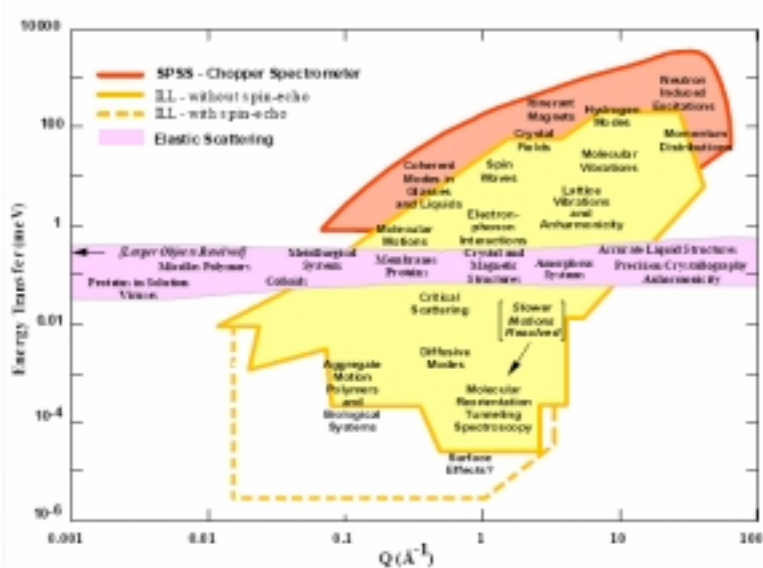


Figure 12. Energy and Wavevector transfer accessible to Neutron Scattering.

## NEUTRON SCATTERING LABORATORY, NATIONAL NUCLEAR ENERGY AGENCY BATAN-INDONESIA

Indonesia has also contributed in the neutron scattering activities, especially in the Asia-Pacific region. The multipurpose reactor G.A. Sywabessy of the National Nuclear Energy Agency (BATAN), Indonesia, which is located at Puspptek Serpong produces also a neutron thermal for the neutron scattering activities. The Neutron Scattering Laboratory at BATAN was officially opened by the President of the Republic of Indonesia in 1992. This laboratory was facilitated by seven neutron scattering instruments: Diffractometer for residual stress measurement (DN1-M/RSM), Four-circle diffractometer /texture diffractometer (DN2), Triple axis spectrometer (SN1/TAS), Neutron radiografi facility (NR1/NRF), Small angle neutron scattering spectrometer (SN2-SMARter), High-resolution small angle neutron scattering spectrometer (SN3/HRSANS) and High-resolution powder diffractometer (DN3/HRPD). The first four instruments were installed at the experimental hall of reactor (XHR), while the last three were installed at the neutron guide hall (NGH). The NGH is connected by a tunnel which comprises two neutron guides transferring neutrons from the G.A. Siwabessy reactor to the instruments in the neutron guide hall. The lay out of the neutron scattering instruments is shown in Figure 13.

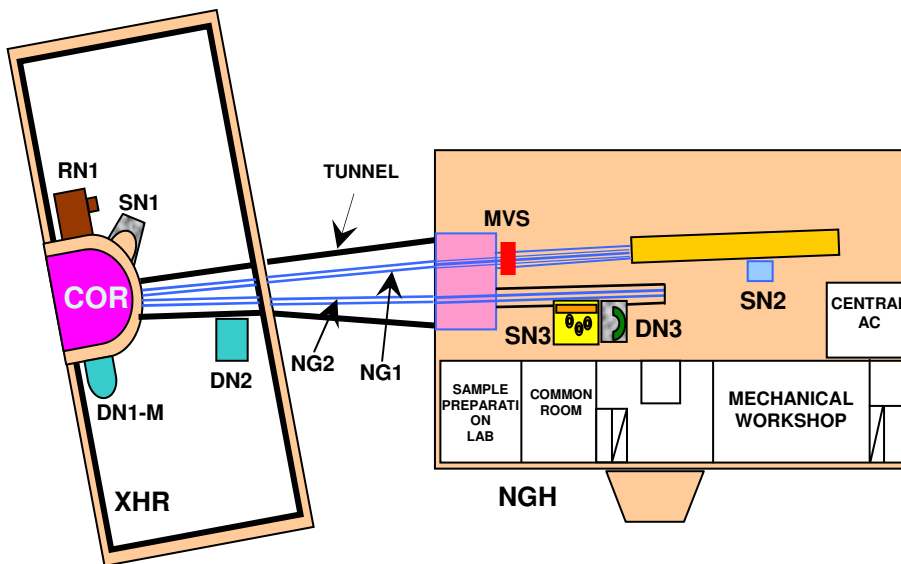


Figure 13. The lay out of the neutron scattering instruments at the Neutron Scattering Laboratory, BATAN, Indonesia.

One of the best spectrometer at NSL is a SANS spectrometer, which was installed at the end of the 49 m long neutron guide tube (NG1) and located in the Neutron Guide Hall (NGH) to benefit from low background environment. A slot type mechanical velocity selector having minimum and maximum rotational speed of 700 and 7000 rpm is used to monochromatize the neutrons beam coming out from the G.A. Siwabessy Reactor. The selector produces neutron wavelengths of 3 – 6 Å and covering the scattering vector of  $0.002 < Q < 0.6 \text{ \AA}^{-1}$  related to size range of 10 – 3000 Å.

The SANS spectrometer consists of a mechanical velocity selector, an 18 m long collimator system comprises four sections of movable guide tube and one section of fixed non-reflecting tube. Another 18 m long tube accommodates a 128 x 128 channels of  $^3\text{He}$  two-dimensional position sensitive detector (2D-PSD) made by RISØ. The detector can be moved continuously from 1.3 m to 18 m from sample position and can also be shifted in the lateral direction by 0.1 m to increase the Q range. Variation of collimator length and sample-to-detector distances (SDD) are fully computer controlled. A photo of SMARTer (SANS Spectrometer) installed in the Neutron Scattering Laboratory at BATAN Serpong, Indonesia is shown below.



Figure 14. A photo of BATAN SANS spectrometer.

A SANS technique has emerged as the advanced technique in research of nano and microstructures in materials and life science, i.e. nano and biotechnology. This technique is applied to solving the molecular morphology in the relevant length scales, i.e. 10-1000 Å of wide range of substances such as alloys, ceramics, polymers, liquid crystals, micellars solutions, colloid suspensions, and also biology materials, e.g. proteins, liposomes etc. The SMARTer has been characterized and calibrated using several standard samples and utilized on alloys, ceramics and solid state polymers and solutions, as well as soft condensed matter studies in order to investigate the size, morphology, conformation, surface structure as well as magnetic structures. The users of SMARTer come from local, regional and international collaborations. Performance of SANS instrument at NSL-BATAN is comparable with the other world-class neutron instruments, such as in JAEA, Japan and ANSTO, Australia.

Table 2. Instrumental parameters of the SANS instrument at NSL-BATAN.

**Instrumental parameters:**

Source	: 30 MW research reactor
Monochromator	: Mechanical velocity sector with variable speed
Incident wavelength range	: 0 – 6 Å
Wavelength resolution( $\lambda/\Delta \lambda$ )	: 10 -20 %
Source-sample distance	: 1.5, 4, 8, 13 and 18 m.
Collimation	: Circular pinholes: 30, 20, 14, 10, 7 and 5 mm.
Max flux at sample position	: $4 \times 10^6$ neutrons $\text{cm}^{-2} \text{s}^{-1}$
Sample size	: >5 mm in diameter
Effective Q range	: 0.002 – 0.6 Å <sup>-1</sup> .
Detector	: 128 x128 <sup>3</sup> He two-dimensional position sensitive detector
Ancillary equipment	: Small heater up to 373K, External magnetic field 1T and Furnace.

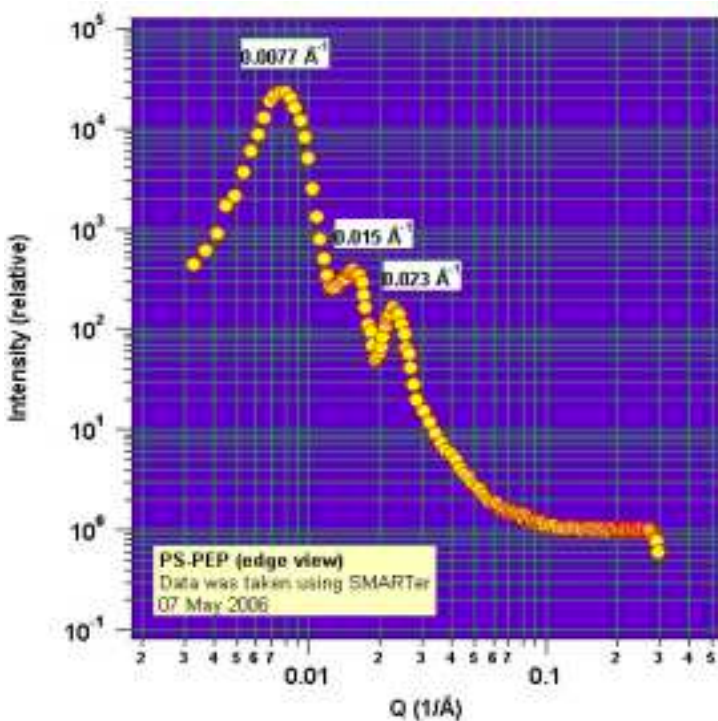


Figure 15. Experiment on solid state polymer sample [14].

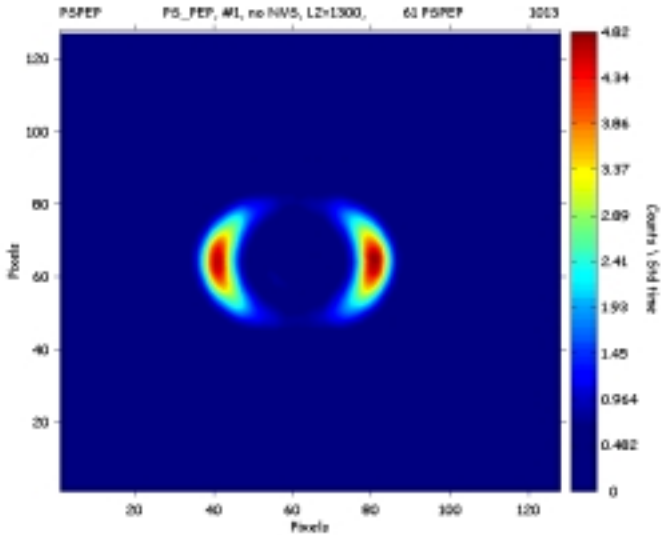


Figure 16. Scattering pattern from Block Copolymer [14].

Figures 15 and 16 show a PS-PEP (polystyrene-block-polyethylene-alt-propylene) and block copolymer film sample, respectively. The result shows an anisotropic scattered pattern, which corresponds to the preferred orientation in lamellar structure. The peak at  $Q = 0.0077, 0.015$  and  $0.023 \text{ \AA}^{-1}$  appeared noticeably and those peaks associate to a distance about  $820 \text{ \AA}$  of the separated phase from two kinds of polymere chain blocks, polystyrene and poly (ethylene-alt-propylene). The second order peak at  $Q = 0.015 \text{ \AA}^{-1}$  can only be observed clearly by applying 20' double-fan mode sectoral radial averaging for subtracting the anisotropic scattering. [14,15].

## CONCLUSION

The promising applicability of neutron scattering to ever-widening fields of scientific study is exciting. More than ever before, the neutron-scattering community is learning how to use the strengths and unique capabilities of different facilities and technologies to their best advantage. The close proximity means that users can exploit the advantages of pulsed and steady-state neutron sources located on the same campus. The goal is to create a haven for neutron science through state-of-the-art facilities, instrumentation, data analysis, and technology development.

With sustained investment in operating and improving existing and new neutron sources, researchers from biology, complex fluids, high-pressure physics, and other fields are increasingly likely to take advantage of the



increased fluxes and instrumentation at the various facilities. Of even greater significance is the scientific impact that cumulative gains in instrument performance are likely to have:

## ACKNOWLEDGEMENT

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