

Pulsed Neutron Scattering for the 21st Century

The large neutron fluxes produced in next-generation spallation facilities are likely to soon find expanded application in fields as diverse as biology, polymers, complex fluids, and geophysics.

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A negatively charged hydrogen ion accelerates down a linac to nearly a billion electron volts—90% of the speed of light—and punches through graphite foil that strips off the ion's two orbiting electrons. The resulting proton enters a ring where it and other protons are stored and accumulated into pulses that are fired at 60 Hz toward a vessel of liquid mercury. In a process known as spallation, the protons collide with atomic nuclei in the heavy metal and knock out short, intense pulses of neutrons. Those neutrons are then guided through as many as 24 beamlines to the myriad instruments and detectors used for experiments.

That's the vision behind the Spallation Neutron Source (SNS), a \$1.4 billion facility nearing completion at Oak Ridge National Laboratory (ORNL). Currently in its testing phase, the facility is expected to produce the most intense pulsed neutron beams in the world, with each pulse yielding neutron fluxes estimated at 20 to 100 times the peak intensity obtainable from fission reactors. Materials of ever-increasing complexity are key elements of today's technologies and underpin the world's industrial and economic development. Consequently, the spallation source will surely find broad applicability in fields far beyond the condensed matter systems to which neutron scattering has traditionally been applied.

Why neutrons?

An ongoing need exists for tools that reveal the microscopic origins of materials' physical, electrical, magnetic, chemical, and biological properties. Beams of x rays and electrons interact strongly with a material's electrons. With x rays, the interaction is electromagnetic; with electrons it is electrostatic.

The neutron's fundamental attributes make it a unique and complementary probe. 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. 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.

Using detectors that consist of highly absorbing nuclei, one can measure the angles at which the neutrons scatter—either elastically or inelastically—and the time at which they are detected. With those data, researchers can infer the momentum and energy dependence of the scattering cross section, a quantum mechanical quantity proportional to the relative contribution of an atom to scattered intensity. The information provides details about the structure and dynamics of materials ranging from liquid crystals to superconducting ceramics, from proteins to plastics, and from metals to micelles to metallic glass magnets.

Although neutrons scatter from nuclei and magnetic moments, and in some cases are directly absorbed, generally they penetrate into the bulk of most materials. That penetration power makes them useful for in situ studies in complex sample environments and for nondestructive measurements in engineering components.

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. Because large biological molecules contain numerous hydrogen atoms, the best way to see part of a biomolecule is through isotope substitution—replacing its hydrogen atoms with deuterium atoms. Conveniently for crystallographers, the nuclei of those atoms scatter neutrons very differently. Using a technique called contrast variation, scientists can highlight different types of molecules, such as a nucleic acid or a protein in a chromosome, and glean independent structural information on each component within a macromolecular complex (see *PHYSICS TODAY*, November 2003, page 17).

Similar techniques can be used to study polymers and other types of hydrogenous materials or to selectively mask portions of complexes in solution by varying the ratio of H to D in the solvent or in subunits of a larger system.

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 is proving equally useful in the study 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.

For a number of chemical elements, figure 1 compares the x-ray scattering cross sections, which scale with the

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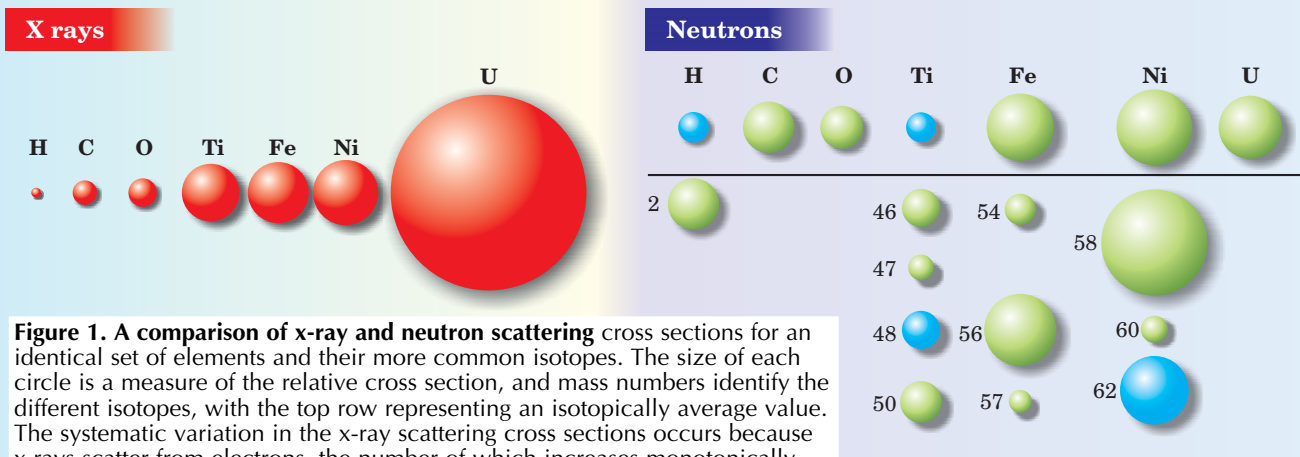


Figure 1. A comparison of x-ray and neutron scattering cross sections for an identical set of elements and their more common isotopes. The size of each circle is a measure of the relative cross section, and mass numbers identify the different isotopes, with the top row representing an isotopically average 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.

square of the number of electrons, with neutron scattering cross sections of those same elements and their more common isotopes. The absence of a systematic variation in neutron cross section across the periodic table is a testament to the complementary information that neutrons provide.

Some nuclei have negative scattering lengths, which can further enhance contrast between nuclei. Negative scattering length—the square root of the cross section—results in a 180° phase shift in addition to the neutron's deflection. The phenomenon affects how neutrons scattered from some nuclei in a material interfere with the neutrons scattered from different nuclei.

Furthermore, the neutron's magnetic moment is large enough that the scattering of neutrons from unpaired electron spins and orbital angular momentum in a material is comparable to scattering from nuclei. That property makes magnetic-structure and dynamics measurements straightforward. Neutron beams can be polarized, and an analysis of the polarization dependence of scattering can further refine the understanding of moment orientations within a material and can separate magnetic from nonmagnetic scattering.

Before researchers can perform any neutron scattering experiments, however, 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.

Passing the neutrons through a cryogenic bath of liquid hydrogen brings them into thermal equilibrium at yet lower temperatures—below 10 meV—and at wavelengths longer than about 3 Å. Those cold neutrons are helpful in the study of systems with longer length scales or those with slower dynamics, including polymers and proteins.

Neutron sources

James Chadwick discovered the neutron in 1932.¹ Two research groups in Europe demonstrated the technique of neutron diffraction soon thereafter.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 performed their pioneering elastic and inelastic diffraction experiments in the 1940s and 1950s and shared the 1994 Nobel Prize for it. Figure 2 shows the evolution of performance among continuous and pulsed neutron sources. Although the scientific utility of reactor-based neutron sources has continued to increase, largely through improvements in instrumentation and neutron optics—the engineering of various solids or multilayers to reflect, focus, and steer the beam—no significant increase in the flux of neutrons from fission reactors has been achieved since the late 1950s.

The limitation on thermal-neutron flux stems from the engineering realities of removing from the core of the reactor the energy released during fission. The energy released per neutron ejected during the spallation process, however, is about an order of magnitude lower than for fission. That efficiency in neutron production prompted researchers in the 1950s to consider using a high-flux spallation source to produce nuclear materials. But it wasn't until construction of the Neutron Science Laboratory (KENS) at Japan's High Energy Accelerator Research Organization (KEK) in 1980 and the Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory (ANL) a year later that the research community saw the first accelerator-based, pulsed spallation sources specifically dedicated to neutron scattering.³ The table on page 47 shows the spallation sources that have operated to date and those under construction.

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 (see the article by Roger Pynn and Brian Fender in *PHYSICS TODAY*, January 1985, page 46). Since then, the US installed a cold source, "guide hall," and associated instruments at what is now the NIST Center for Neutron Research in Maryland.⁴

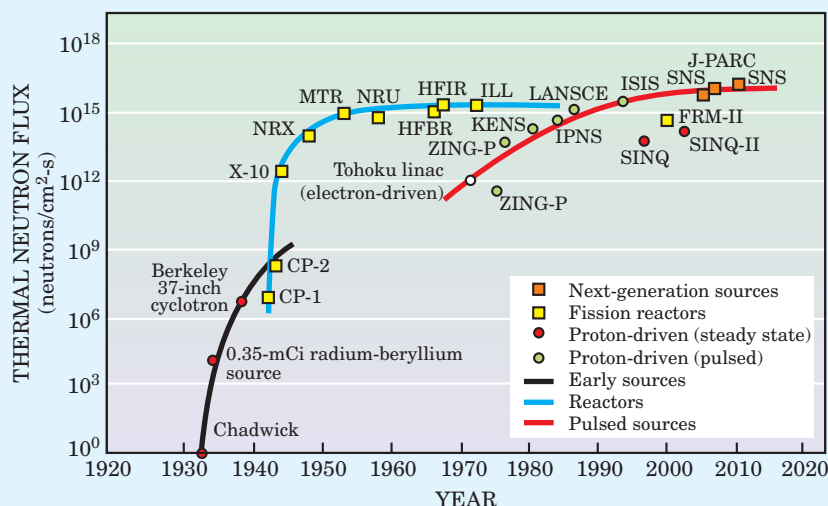


Figure 2. Historical development of thermal-neutron sources, both continuous and pulsed. The plot of peak neutron flux neglects variations in the efficiency of beam extraction and instrumentation, both of which can have a significant impact on performance. The earliest neutron sources were based on radioactive 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 two SNS data points refer to the peak flux of neutrons expected this year and the peak expected in 2011 after a power upgrade. (Figure updated from ref. 9.)

In the mid-1990s, following advice from a series of National Academy of Sciences reviews and advisory committees, the US Department of Energy initiated and later approved the development of the SNS at ORNL.

The SNS

What largely distinguishes the SNS from other major facilities is the distributed design and construction partnerships that contributed to its development. National laboratories across the country designed and procured different systems, while ORNL installed and integrated those systems. The various components that constitute the SNS include the following:

- ▶ a front end composed of a hydrogen-ion source, a radio-frequency quadrupole that guides and accelerates the ions, and a medium-energy beam transport system, developed at Lawrence Berkeley National Laboratory;
- ▶ two linear accelerators, one a normal-conducting accelerator designed by Los Alamos National Laboratory, the other a superconducting accelerator designed at Thomas Jefferson National Accelerator Facility;
- ▶ an accumulator ring developed by Brookhaven National Laboratory;
- ▶ a mercury target, civil construction, and integration at ORNL;
- ▶ and an initial instrument suite designed and developed by ANL.

Figure 3 shows the SNS layout. Upon completion, the SNS will be operated by ORNL and available to visiting researchers with peer review serving to evaluate proposed experiments. The annual operating budget is expected to be \$160 million.

At 1.4 MW, the SNS has approximately eight times the beam power of ISIS in the UK—currently the world's leading pulsed spallation source. The SNS provides space for 24 beamlines. Seventeen instruments for those lines are in various stages of development or design. If approved, funding in the 2007 budget will bring the total number of instruments to 20. Because many of those instruments represent improvements to the state of the art, the SNS will make it possible to take measurements of greater sensitivity, at higher speed, with higher resolution, and in more complex sample environments than has ever been possible at existing facilities.

The design specifications involved several technical challenges. The SNS incorporates the first superconducting pulsed proton linac, an addition that vastly increases the accelerator's electrical efficiency and adds flexibility to its operation. And the use of mercury as a target in a pulsed environment is a first. Liquid metals don't suffer the radiation damage that solids do and are better able to dissipate heat and withstand the shock of high-energy pulses.

The initial experience with and testing of the superconducting linac and accumulator ring have been

promising. Incorporating the superconducting linac into the design provides a natural upgrade path to higher beam power as beam energy increases. The DOE has given initial approval and funding to begin work on the upgrade, and additional beam power could be available by 2011. In the longer term, by the middle of the next decade, a second target station, optimized for long-wavelength neutrons, may be built.⁵

International context

Worldwide, accelerator-based spallation sources of neutrons are much less numerous than reactor-based ones. To the list of pulsed-neutron sources in the table, add the continuous Swiss Spallation Neutron Source (SINQ) facility at the Paul Scherrer Institut in Switzerland. From a scattering-instrumentation point of view, that facility has the character of a medium-flux reactor, although it is based on a spallation target driven by a high-current proton cyclotron.

The SNS at Oak Ridge 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.⁶ Although efforts in Europe to build a next-generation spallation source have not yet led to a funded project,⁷ 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.

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.

Pulsed Spallation Neutron Facilities

Source,* turn-on date	Proton energy, Current	Pulsing frequency (Hz)	Target material	Status
ZING-P, ANL, US 1974	300 MeV 100 nA	30	Lead	Shut down 1975
ZING-P', ANL, US 1977	500 MeV, 3 μ A	30	Tungsten, uranium	Shut down 1980
KENS, KEK, Japan 1980	500 MeV 7 μ A	20	Tungsten, uranium	Operating until 2006
IPNS, ANL, US 1981	450 MeV 15 μ A	30	Uranium (including enriched U)	Operating
ISIS, Rutherford, UK 1985	800 MeV 200 μ A	50	Tantalum, tungsten, uranium	Operating
MLNSC, LANL, US 1986	800 MeV 100 μ A	20	Tungsten	Operating
IN-6, INR Troitsk, Russia, 1998	600 MeV up to 500 μ A	50	Tungsten	Operating intermittently
SNS, ORNL, US 2006	1 GeV 1.4 mA	60	Mercury	Testing phase
J-PARC, Japan, 2008	3 GeV 333 μ A	25	Mercury	Under construction

*Source acronyms refer to the Zero-gradient synchrotron Intense Neutron Source-Prototype and Intense Pulsed Neutron Source, both at Argonne National Laboratory; the Manuel Lujan Jr. Neutron Scattering Center at Los Alamos National Laboratory; the Institute for Nuclear Research in Russia; the Japan Proton Accelerator Research Complex; and the Spallation Neutron Source at Oak Ridge National Laboratory.

ties. Of even greater significance is the scientific impact that cumulative gains in instrument performance are likely to have: Improved neutron optics, new types and numbers of detectors, sophisticated sample environments, and powerful software for collecting and visualizing data will all combine to heighten the opportunities for researchers in various fields who might otherwise find it impractical to add neutron scattering to their methods of solving problems.

Scientific opportunities

The various neutron-scattering instruments installed on the 24 beamlines served by the target station will determine the range of topics that SNS can address. Each in-

strument 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.

With a time-of-flight source like SNS, the challenge is to design and build instrumentation that fully counts useful neutrons—those in the energy range of interest—during the entire interval between pulses. Each pulse contains neutrons of different wavelengths, and therefore of different velocities, that arrive at the detector at different times. At pulsed sources worldwide, the trend, driven particularly by developments at ISIS in the last 10 to 15 years, has been to use incident flight paths and beam optics that optimize the dynamic range and flux. Detector arrays that cover very large solid angles then maximize counted neutrons. That is, instead of using a discrete set of time-of-flight detectors, researchers cover the walls of the various spectrometers with them. That approach has led to a growing number of instances in which the instrument performance is well matched to the peak neutron flux, a metric that is well served by a high-performance pulsed neutron source like SNS.

Through a peer-review proposal system, SNS has allocated 17 beamlines so far. Information in each of those instruments can be found on the SNS website at http://www.sns.gov/users/instrument_systems/index.shtml. The box on page 48 pictures the initial seven of those instruments and the following short descriptions outline the scientific opportunities they provide.

The backscattering spectrometer is intended for study of atomic-scale dynamics with high energy resolution (2.5 to 10 μ eV) and up to 18-meV energy transfer. To reach that energy resolution, the spectrometer combines an 84-meter incident flight path in front of the sample with high wavelength selectivity from backscattering off an array of silicon crystals surrounding the sample.

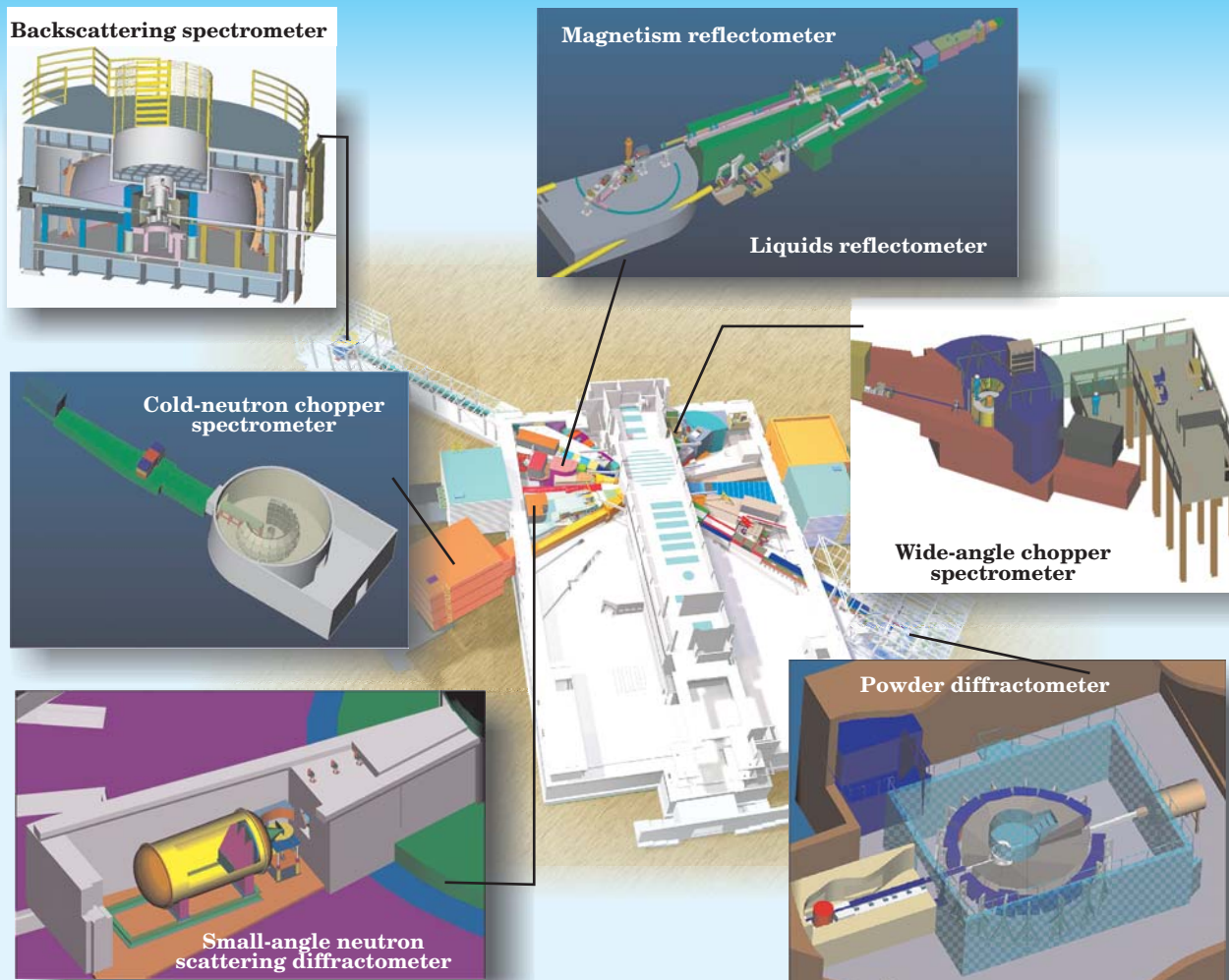
The unique combination of dynamic range and energy resolution, together with high intensity, shrinks the required neutron-counting time to 30–100 times less than that required when existing instruments are used. Experiments can thus better resolve macromolecular dynamics of biomolecules and polymers, solvent dynamics of micelles at nanometer scales, the diffusion of hydrogen within metallic nanoparticles and ions within conductors, and the dynamics of films and lubricating molecules, to list a few examples.

The beam intensity will be high enough to let researchers study even very small sample volumes, weakly scattering isotopes, and small magnetic moments while changing temperature, magnetic field, or other variables. At high energy transfer, the backscattering spectrometer will provide a unique view of low-energy magnetic



Figure 3. Layout of the Spallation Neutron Source, from an aerial photo taken in September 2005. The facility, scheduled for completion this June, is located north of the main Oak Ridge National Laboratory campus.

Overview of the Initial Spallation Neutron Source (SNS) Instruments and Techniques



This composite image illustrates the first seven instruments out of an eventual 24 that will be built on the initial SNS target station. (A second target station is planned about 10 years down the road.) Engineers at the facility plan to bring instruments into service at a rate of about two per year, with the initial instruments ready for user operation in 2008 and the balance built over the subsequent years. Each instrument exploits neutron scattering using different geometries and energy ranges to answer specific questions or explore particular materials of interest. The challenge is to measure the intensity of scattered neutrons as a function of momentum and energy transferred to the sample. That intensity is proportional to the static and dynamic structure factors for the materials being studied—where the atoms are and how they move.

In the case of the backscattering spectrometer, the entire flux of neutrons scatters from the sample in the center of an evacuated tank and is then reflected at nearly 180° from a spherically shaped silicon-crystal array onto a detector. That silicon analyzer crystal selects a single wavelength that gets backscattered onto the detector.

Powder diffraction experiments similarly involve a sample irradiated with neutrons having a wide energy range. But all the scattered neutrons are recorded, and their arrival times monitored, in banks of detectors located at a wide variety of angles around the sample. The neutron's time of flight is proportional to its wavelength, and the wavelength proportional to the spacing between atomic planes, so measurements of

neutron scattering can be plotted against time of flight, wavelength, or lattice spacing.

The chopper spectrometers—both wide-angle and cold-neutron varieties—invert what happens in backscattering. Each exploits a subset of the neutron flux—only those in a narrow-energy window. Mechanical choppers are phased to spin open and closed with the neutron pulse and thus pass only neutrons with a particular velocity. The monochromatic neutrons hit the sample and lose or gain energy on scattering. The time of flight to the detectors indicates the extent of energy transfer. What distinguishes the two spectrometers are the energies of neutrons.

The magnetism and liquids reflectometers share the same multichannel shutter that opens the beamline to neutrons. For studying magnetic surfaces and multilayers or liquid surfaces, the beam must be scattered at glancing incidence. Preventing vibrations that would create ripples or otherwise interfere with measurements is crucial in liquid reflectometry.

To measure diffraction from large molecules such as proteins and polymer blends, the transferred momentum (and thus scattering angle) must be small and the neutron wavelength large. But unlike the reflectivity configuration, the neutrons do not hit at glancing incidence in so-called small-angle neutron-scattering experiments. Rather, neutrons are sent through a pinhole to the sample, and the scattered outgoing wave of neutrons passed on to a large-area detector in an evacuated tank.

excitations in correlated electron systems and reveal subtle effects in lattice dynamics and electron–phonon coupling.

Reflectometers scatter neutrons at glancing incidence to probe surfaces, interfaces, thin films, and multilayers. SNS will have two such instruments: one equipped for polarized neutrons and the study of magnetic and chemical-density profiles, the other designed to probe density profiles normal to the surface of a liquid. Both will have data rates 10 to 100 times higher than the best existing instruments and be able to measure reflected intensity at levels 10 times smaller than can currently be resolved; the effect is an enhanced sensitivity at shorter length scales because reflected intensity falls off rapidly as a function of inverse length scale.

Both instruments should find application in studies of phase separation in polymer films, surfactants at interfaces, protein adsorption, nucleation and growth of structured surfaces, and magnetic domains and patterned structures of magnetic dots. The more traditional layer-averaged measurements are likely to be used to study the formation of magnetic moments in thin films, giant and colossal magnetoresistance, molecular magnets, complex fluids under flow, reaction kinetics, and critical phenomena in fluid systems.

Extended Q-range small-angle neutron scattering (EQ-SANS) is used for low-resolution studies of materials with long length scales. The low scattering angles and long neutron wavelengths probe structures as large as hundreds of nanometers. The SNS's broad range of neutron wavelengths is well suited for a wide variety of materials, including biological molecules, polymers, and colloidal systems. Researchers in life sciences can scatter neutrons from solutions of proteins, DNA, and other biological complexes; chemists can probe polymer and colloidal systems such as block copolymers, micelles, and aerosols; materials scientists can investigate nanoparticles, crystallization, precipitation, and phase-separation phenomena; and Earth scientists can learn more about pore structure and contaminant uptake in soil and about the fractal structure of rocks.

Powder diffraction probes the atomic structure of powdered crystalline samples. The technique has been a particular strength of pulsed-neutron scattering because of the favorable matching of wavelength range and resolution that can be obtained for typical accelerator, moderator, and instrument parameters. The SNS instrument uses a large, contiguous detector array and will be an extremely flexible and versatile general-purpose diffractometer useful for a wide range of structural studies.

Its geometry makes the powder diffractometer especially suited to studies of a material's response to applied changes in temperature, pressure, or chemical environment, and for elucidating magnetic and nonmagnetic crystal structures with unprecedented precision and speed—even in cases where single-crystal data may not be available. Magnetic systems, including high- T_c superconductors, charge- and orbital-ordering transitions, perovskites, and geometrically frustrated systems, and nonmagnetic systems, including zeolites, fuel-cell materials, gas hydrates, minerals, and cements, are among the areas of interest.

Chopper spectrometers are inelastic scattering instruments that use rotating mechanical gates to select a particular incident neutron energy. The time-of-flight record determines the scattered neutron energy. The cold-neutron chopper spectrometer (CNCS) covers the dynamic range of interest in complex fluids—dilute protein solutions and biological gels, for example—excitations of quan-

tum fluids in confined geometries, quantum effects in low-dimensional magnetic systems, dynamics of water and ionized liquids in confined geometries, and the relaxation dynamics of water in cement.

A high-resolution, time-of-flight chopper spectrometer has been optimized to provide a high neutron flux at the sample, the scattering from which is measured through large solid angles. The instrument is designed to measure excitations in materials more efficiently than any existing high-energy (between roughly 40 meV and 1 eV) chopper spectrometers. Typical research topics include studies of vibrational excitations and their relationship to phase diagrams and equations of state of materials, and studies of spin correlations in magnets, superconductors, and materials close to metal–insulator transitions.

With the balance of the instrument suite, which will extend the facility's capabilities to yet longer and shorter length and time scales, the SNS will support a broad range of investigations, ranging from macromolecular diffraction and engineering materials to high-pressure studies. The combined gains of a more intense neutron source and state-of-the-art instrumentation will further broaden the application of neutron scattering beyond its traditional base in condensed matter. There is even a beamline dedicated to the study of the neutron itself; that beamline will address questions beyond the standard model, including whether the neutron actually has an electric dipole moment.

Great expectations

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. Once completed, the SNS will operate two miles from the newly upgraded High Flux Isotope Reactor (HFIR) facility at ORNL. 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.⁸

The Spallation Neutron Source is managed by UT–Battelle LLC, under contract DE-AC05-00OR22725 for the US Department of Energy. The SNS is a partnership of six national laboratories: Argonne, Brookhaven, Thomas Jefferson, Lawrence Berkeley, Los Alamos, and Oak Ridge.

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