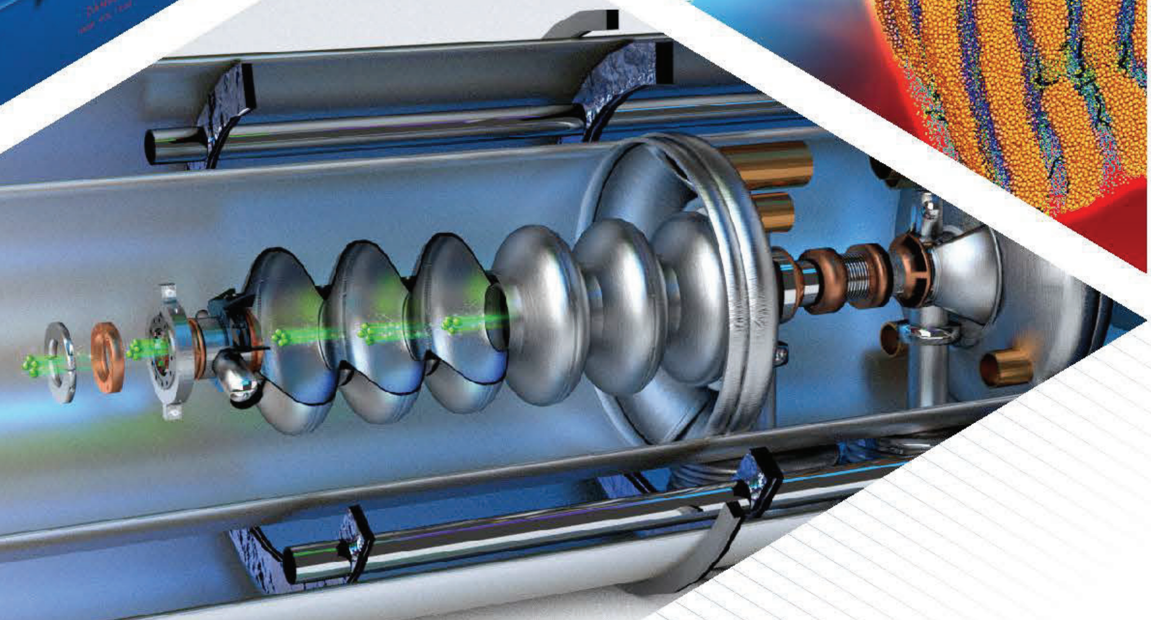
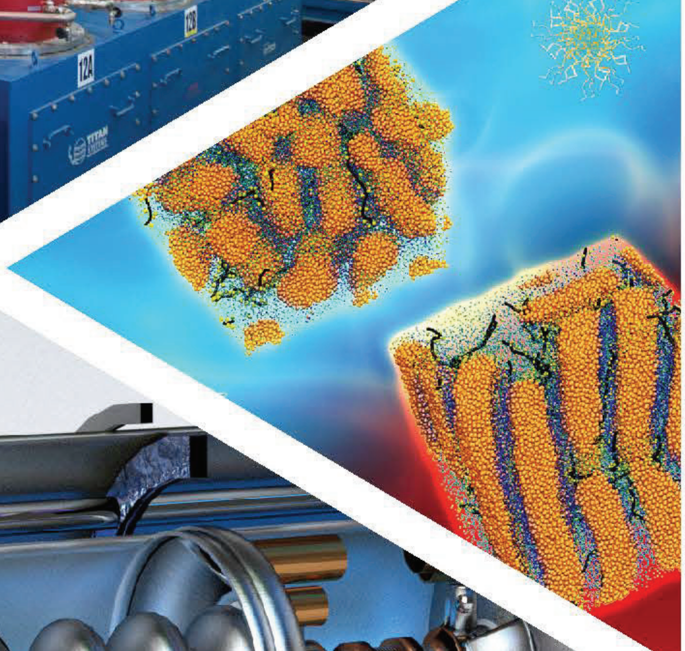
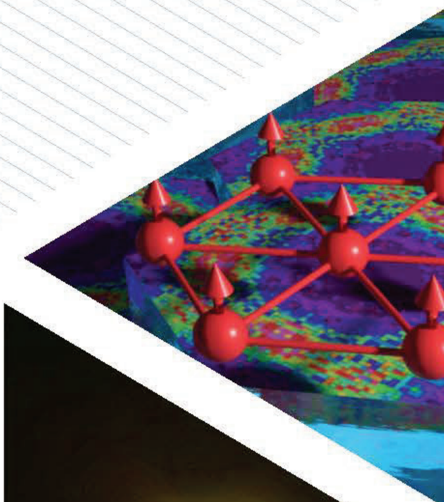
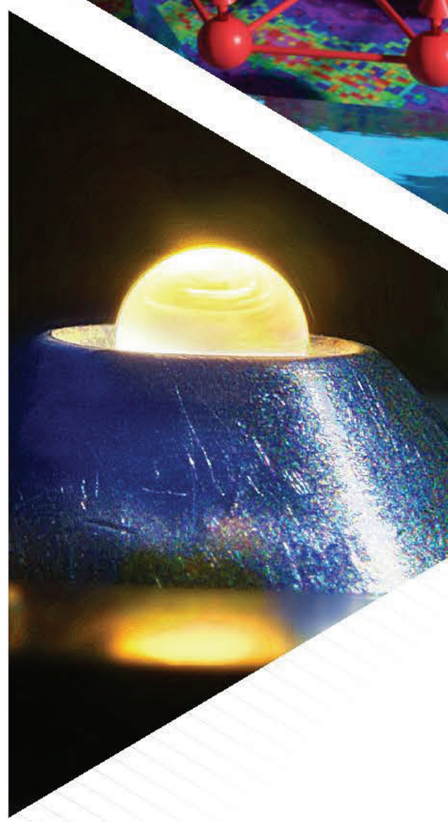
The Science Case for a Proton Power Upgrade

Spallation Neutron Source









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OAK RIDGE
National Laboratory

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PREFACE

Oak Ridge National Laboratory (ORNL) operates and develops the Spallation Neutron Source (SNS) and the High Flux Isotope Reactor (HFIR) as advanced neutron scattering user facilities and centers of innovation and scientific excellence. Providing researchers with access to cutting-edge neutron scattering capabilities enables them to use the unique properties of neutrons to advance scientific discovery and to solve the most challenging technology problems.

Keeping ORNL resources like SNS and HFIR at the forefront internationally is central to ensuring continued US leadership in science and technology. The Proton Power Upgrade (PPU) project, which seeks to increase the neutron flux of beams at the First Target Station (FTS) and to provide the capability to power a future Second Target Station (STS), is critical to maintaining US leadership in neutron scattering and related technologies. PPU will involve upgrading the SNS accelerator complex to double the currently available proton beam power from 1.4 to 2.8 MW. With 2 MW of this delivered to an improved FTS, urgently needed increases in flux are anticipated, which will enable new scientific discoveries in the areas of soft matter, quantum materials, chemistry, functional materials, and biology. PPU will make possible experiments that currently are not feasible or routine such as time-resolved in situ measurements, experiments on smaller or less concentrated samples, and experiments under more extreme environmental conditions.

PPU will also provide a platform for a future STS with the highest peak brightness beams of cold neutrons at low repetition rates in the world. STS in turn will provide unprecedented access to mesoscale and complex matter, complementing the existing and future capabilities of FTS, with its high peak brightness beams of thermal neutrons at high-repetition rates, and HFIR, with its continuous beams with high time-averaged flux. This three-prong strategy will ensure that US researchers have access to leading-edge neutron scattering capabilities to address critical emerging challenges for the foreseeable future.

This pamphlet and a companion piece, *Early Scientific Opportunities at the Second Target Station*, describe some of the exciting new possibilities coming with PPU.

THE SCIENCE CASE FOR A PROTON POWER UPGRADE

The Proton Power Upgrade (PPU) project will add new scientific capabilities to the Spallation Neutron Source (SNS) by increasing the flux of pulsed beams of neutrons at the existing First Target Station (FTS). This will enable new scientific discoveries through experiments that currently are neither feasible nor routine such as time-resolved in situ measurements, experiments on smaller or less concentrated samples, and experiments under more extreme environmental conditions. An additional benefit of the increased flux delivered by PPU is shorter durations for experiments and therefore an increased number of possible experiments. This in turn will lead to increased researcher access to neutron scattering capabilities and an accelerated pace for scientific discoveries that rely on neutrons to provide unique information about the structure and dynamics of matter. PPU will also provide the capability to produce more proton power to drive a future Second Target Station (STS). STS will complement FTS as the world's highest peak brightness cold neutron source with game-changing new capabilities for emerging science.

INTRODUCTION

Scientific breakthroughs rely on the availability of advanced tools and user research facilities. The US Department of Energy (DOE) Office of Basic Energy Science (BES) maintains multiple user facilities at national laboratories across the nation as national resources that researchers from universities, national laboratories, research institutes, and industry can use to conduct experiments that would be impossible to conduct at their home laboratories. Among the user facilities operated by the Oak Ridge National Laboratory (ORNL), SNS and the High Flux Isotope Reactor (HFIR) provide neutron scattering capabilities that have enabled researchers to make scientific discoveries that would not have been possible using other techniques.

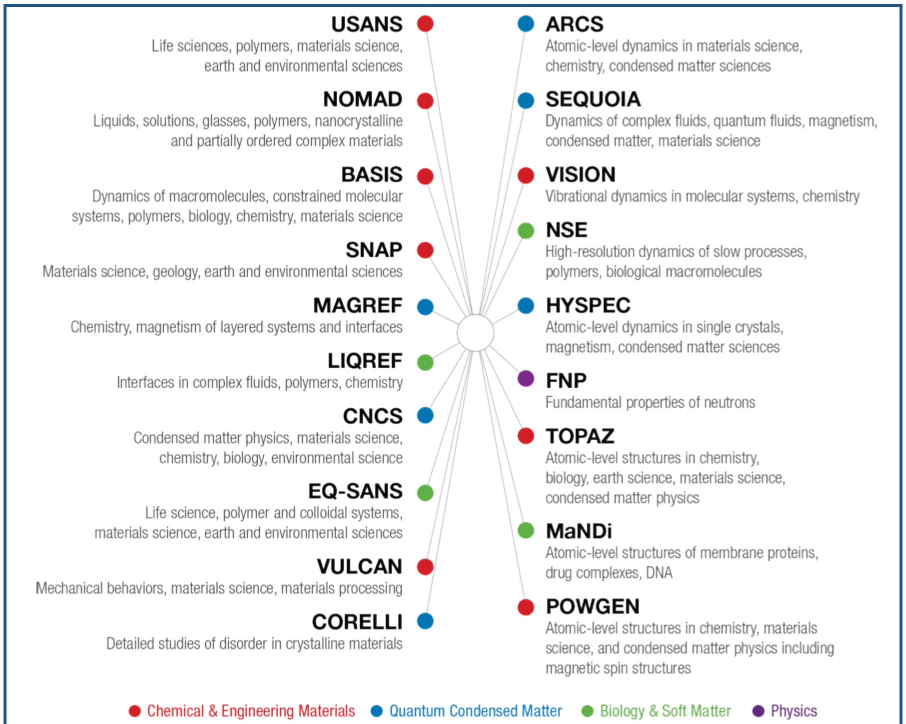

Figure 1. Beamline instruments at the Spallation Neutron Source. (Figure courtesy of Jill Hemman, Neutron Sciences Directorate, Oak Ridge National Laboratory.)

SNS was conceived as a third-generation neutron source with the first short-pulse megawatt-class proton accelerator, which provides beams of pulsed neutrons with unprecedented peak brightness for scattering experiments. The advantage of neutron beams with a pulsed structure is that they enable time-of-flight

(TOF) instruments in which the energy (wavelength) of each scattered neutron is recorded. The suite of existing instruments covers length scales from the atomic (angstroms) to the macroscopic (centimeters) and timescales from tenths of picoseconds to hundreds of nanoseconds (Figure 1). SNS instruments are leading edge, with unique capabilities and performance levels, and they are continually improved to ensure that SNS enables cutting-edge neutron scattering science.

Since the start of the user program in 2007 SNS has seen growth in neutron scattering capabilities and capacity, scientific productivity and impact, and the size and breadth of its user community (see Appendix Note 1). However, it is urgent that neutron scattering capabilities be further expanded to enable researchers to address new scientific challenges that have been identified as important by DOE and the science community (see Appendix Note 2). PPU, which seeks to increase the neutron flux and peak brightness of FTS and provide the capability to power a future STS, is critical to this expansion. The increase in proton power delivered to FTS by PPU will increase the flux of neutron beams by about 40% on average, and the increase in peak brightness of neutron pulses at some energies has the potential to be much higher. These gains will enable new measurements and capabilities that are currently only at the threshold of feasibility. These new measurements and capabilities will also benefit from simultaneous advances in neutron scattering technologies, data analysis, and theory and computation. Additional benefits of PPU include providing broader researcher access to neutron scattering capabilities, increasing the number of experiments possible, and accelerating the pace of scientific discoveries that rely on neutrons to provide unique information about the structure and dynamics of matter.

On the following pages we explore some of the advanced applications that depend on neutrons and will only be possible with PPU.

SOFT MATERIALS

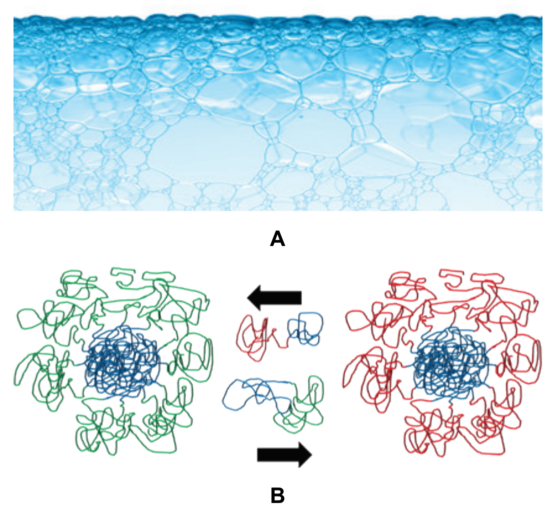

Figure 2. PPU will enable time-resolved neutron scattering studies combined with specific deuteration to characterize the molecular processes responsible for improved industrial performance in materials such as surfactants (A) and n-alkyl-PEO polymers (B). (Figure 3A courtesy of iStock, Getty Images; 3B courtesy of Gregory A. Smith, Oak Ridge National Laboratory scientist.)

Materials that are easily deformed or that contain organic macromolecules are usually referred to as "soft." They can take the form of anything from dilute solutions and soft pastes to rigid polymer composites that are used in the automotive and aviation industries. Everyday examples include plastics made from polymers and detergents made from surfactants (Figure 2A). The chemical diversity of the compounds encompassed by the sobriquet "soft matter" is vast and growing daily due to advances in synthetic chemistry and processing. Covalently bound molecular building blocks such as polymers or surfactant molecules may be assembled into bulk materials having desirable macroscopic properties, thus resulting in nearly endless opportunities for creating materials by design with specific functionalities. It is not only important to study the unperturbed static and dynamic behavior of newly designed soft materials but also to investigate them in situ as part of operating devices or under real-world processing conditions. An equally important aspect of the progress in soft matter is the ever-increasing degree of synthetic control over commodity materials.

By varying characteristics such as molecular weight, tacticity, composition, polydispersity, and branching/topology, it is possible to achieve multifold improvements in material performance.

Neutron scattering is an invaluable tool for studying newly designed soft materials because neutrons are highly penetrating, nondestructive, and particularly sensitive to hydrogen—a key component of most soft materials that is difficult to locate or invisible using other scattering techniques. Furthermore, substituting deuterium for hydrogen greatly changes how samples interact with neutrons, allowing researchers to highlight particular molecules or parts of molecules. Time-resolved neutron scattering studies of preparation processes for and the performance of polymer-based materials and devices is an important new capability that would be enabled by PPU. For instance, n-alkyl-poly(ethylene oxide) (n-alkyl-PEO) polymers have large soluble "matrix" blocks and relatively small insoluble "core" blocks that enable them to form star-like micelles in aqueous solutions. These solutions have rheological behaviors that suggest potential applications in the materials and pharmaceuticals industries. The key to these behaviors is the exchange of polymers between micelles (Figure 2B). Small angle neutron scattering (SANS) combined with specific deuteration is ideally suited to follow this process in situ at a molecular level (Zinn et al. 2012). When the solution of micelles is relatively concentrated the exchange rate is around 1 minute, which matches the time to record a data snapshot from this system on the SNS SANS instrument EQ-SANS. As the concentration or temperature is changed, the exchange rate can increase—to less than 1 minute—and the viscoelastic properties are significantly improved. However, under these conditions the required time resolution is beyond the current capabilities of EQ-SANS. The gains in neutron flux provided by PPU would push EQ-SANS across the threshold into the time-domain that is relevant for characterizing the processes responsible for improved industrial performance properties.

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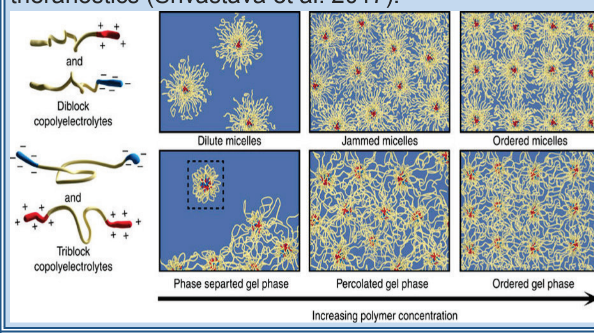
Soft Materials. The sensitivity of neutrons to hydrogen; the possibility of selective deuteration; and the high-penetrating, nondestructive nature of neutrons were used in scattering studies of the assembly of functionalized polyelectrolytes. Diblock polymers assemble into star-like micelles. At low polymer concentrations, discrete micelles remain suspended in the solution. With increasing polymer concentration, micelles jam, thus forming viscoelastic solids, and eventually order in cubic lattice structures. Triblock polymers are expected to form flower-like micelles at extremely low concentrations but instead form gels that phase separate from the solution. This discovery creates the possibility of new applications for gels in tissue engineering, agriculture, water purification, and theranostics (Srivastava et al. 2017).



n-alkyl-PEO polymers are part of a family of soft materials known as "block copolymers." Because of their complex structures, block copolymers have many interesting properties and almost limitless possibilities for use through tuning their architectures. Functionalized block copolymers can be assembled into complex molecular structures with numerous applications, including intelligent fuel cell membranes and biological sensors. Another interesting example is a material consisting of polymers with functional sulfonated polystyrene end blocks and a structural polyethylene middle block (S-SES) that may be capable of serving as a drug delivery device for doxorubicin, an important cancer drug (Figure 3; Chen et al. 2016). Fine-tuning water diffusion in such a device has been proposed as a method for controlling structural changes that would release drugs over the course of a minute. So far, a time resolution of only about 3 minutes has been obtained with the EQ-SANS instrument—inadequate for this application. The extra neutron flux provided by PPU would allow us to cross the threshold into the time domain relevant for the functioning of such assemblies.

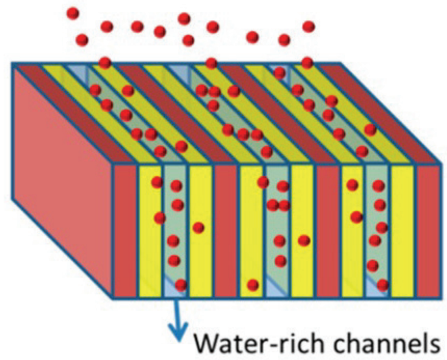


Figure 3. S-SES lamellar membrane of the type envisioned for drug capture and delivery. (Figure from Chen et al. 2016).

PPU would also enable new classes of experiments designed to reveal how to control the self-assembly of block copolymers for surface templates, how to understand protein or lipid organization in membranes, how to understand the response of active films of functional polymers to changing conditions, and how to tune surface properties such as superhydrophobicity by patterning and coating functionalized surfaces. These types of experiments require information on the structure within the plane of surfaces and interfaces. That information can be provided with techniques called grazing incidence diffraction and grazing incidence SANS. These techniques are not routinely used at SNS because of flux limitations; an increase in neutron flux such as that provided by PPU is required for routine use. As another example, the reflectivity curve from a block-copolymer film can currently be obtained in about 30 minutes. With the increased flux from PPU a region of that curve could be monitored as an electric field is applied with a time-resolution of about 1 minute.

Soft materials are integral to a wide variety of products used in our daily lives, yet the behavior of these materials during manufacture is not fully understood. Consider, for example, polymers under flow, an industrially relevant processing condition that lies between the initially disordered structure and the final crystalline material. The transition between states during manufacture is very challenging to study because it takes place in a very narrow range of conditions during processing. PPU will enable more detailed studies of materials near this transition, thereby shedding new light on the polymer crystallization process that is important to many industries and products. Similarly, the additional flux provided by PPU will make it possible to more accurately map other structural transitions in materials such as phase separations in novel composite materials and temperature-dependent transitions in microemulsions used in the food and pharmaceutical industries.

Liquids and solutions are of prime importance in nearly every facet of geology, chemistry, and biology. Experimental probes of the detailed intermolecular forces in liquids and solutions are a critical missing "ingredient" in the quest for quantitative theories. Beyond their sensitivity to light atoms such as hydrogen, another advantage of neutrons for studying the structure and dynamics of liquids is that they do not create the local changes in pH and free radicals that are associated with the absorption of x-rays. However, neutron scattering typically requires large quantities of concentrated sample to increase the weak scattering signal, and this can be a significant limitation. For example, with polyelectrolytes in solution it is crucial to keep the (molecular weight–dependent) concentration below the threshold beyond which they start to aggregate and change functionality. PPU would lower the required sample concentrations, extending the experimental parameter space to encompass a substantially larger part of the functional parameter space (Figure 4).

QUANTUM MATERIALS

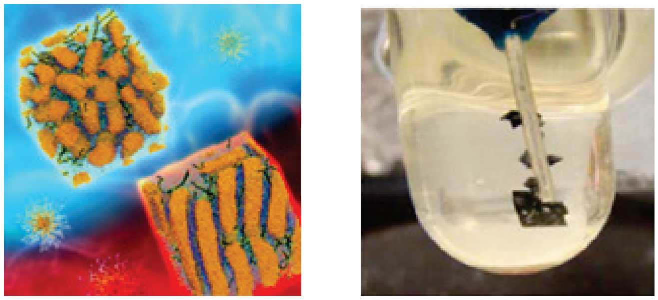


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Figure 4. The increase in neutron flux provided by PPU will enable use of (A) solutions with lower sample concentrations, which is important for mapping the phase diagram of polymers and surfactants, and (B) smaller samples, which is important for enabling the study of newly synthesized materials such as new organic superconductors or biological complexes that are difficult to grow to large crystals. (Images courtesy of Jill Hemman and Genevieve Martin, Neutron Sciences Directorate, Oak Ridge National Laboratory.)

As explained in a recent *Nature Physics* editorial (*Nature Physics* 2016), the term "quantum materials" evolved from the focused effort to understand the inherent complexity of high-temperature superconductors combined with the realization of the importance of topology in materials. These materials exhibit a variety of novel phenomena driven by quantum fluctuations, quantum entanglement, and the underlying topology of the resulting quasiparticles. There is increasing recognition of their potential for next-generation applications in energy and information: quantum materials may enable the development of computers that are exponentially more powerful than what we use today; new energy-efficient, high performance electronic devices like cell phones; and lossless power transmission.

Neutron scattering has long been an essential tool in condensed matter physics, mainly because of the magnetic moment of the neutron and the energy scale of thermal and cold neutron beams. The high peak brightness of SNS, together with TOF techniques, has made it possible to rapidly survey large 4-D volumes of momentum-energy space, providing a powerful tool for identifying and studying emergent properties arising from collective effects in quantum materials. PPU will make specific experiments feasible that are currently only marginally possible. Examples include measurements in high pressure, high magnetic fields and inelastic neutron scattering (INS) using polarization analysis. The need to go to higher pressures and higher magnetic fields to explore quantum materials requires much smaller sample sizes owing to the physical limitations of sample environment designs [e.g., diamond-anvil cells (DACs) for high pressures and pulsed electromagnets for high magnetic fields (> 30 tesla)]. By delivering a higher neutron flux, PPU will make use of these smaller sample sizes possible (Figures 4 and 5).

For instance, current experiments using pulsed high magnetic field magnets at SNS have a duty cycle that allows for only ~1 second's worth of actual data to be collected over a 3-day period. To properly explore magnetic phase diagrams, experiments such as these need to be more efficient. Similarly, the sample volumes in DACs are necessarily small, typically much less than 1 mm^3, making INS at high pressures a challenge. Nevertheless,

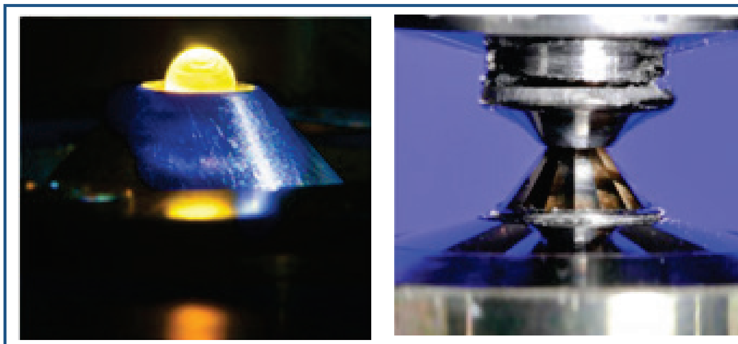

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Figure 5. The increase in neutron flux provided by PPU will enable the investigation of more extreme conditions such as (A) higher temperatures with aerodynamic levitators to study alloys and liquids and (B) higher pressures, which is important for in situ studies of materials synthesis and for exploring the phase diagrams of quantum materials such as superconductors and multiferroics beyond current limitations. (Images courtesy of Jill Hemman and Genevieve Martin, Neutron Sciences Directorate, Oak Ridge National Laboratory.)

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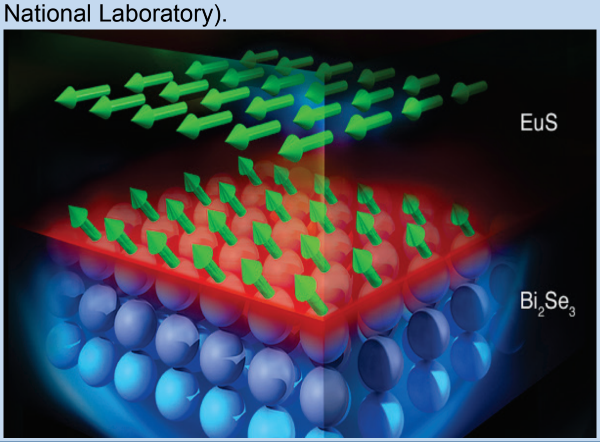
Topological Insulators. A unique property of topological insulators is that electrons can flow on the surface without dissipation while the bulk of the material serves as an electrical insulator. To develop practical devices based on this property, magnetism can be introduced without disturbing the bulk insulating properties by interfacing the surface of the topological insulator with a layer of ferromagnetic insulating film. The sensitivity of neutrons to magnetism and their innate ability to pass through materials in a nondestructive fashion was used in a polarized neutron reflectometry study to detect such a hidden magnetic signal. The discovery promises new opportunities for next-generation electronic and spintronic devices such as improved transistors and quantum computing technologies (Katmis et al. 2016). (Image credit: Jill Hemman, Neutron Sciences Directorate, Oak Ridge National Laboratory).



major advances in increasing the sample volume of DACs have been made at SNS, as recently demonstrated on the vibrational spectrometer instrument, VISION. By combining the higher flux provided by PPU with recent SNS advances in DACs (Figure 5B) and pulsed high field magnet technologies, researchers will be able to explore previously unattainable phase space for neutron diffraction and spectroscopy with the possibility of discovering new material behaviors and phenomena.

A specific science problem in this realm relates to the new world record superconductor, hydrogen sulfide (H_2S), which has a transition temperature above 200 K at 150 GPa. SNS DACs have achieved record pressures for neutron scattering experiments of near 100 GPa. The crystal structure evolution of H_2S (or D_2S) is known to have several phase transitions as a function of pressure, but due to the extremely weak x-ray scattering from hydrogen, the crystal structures of these high-pressure phases are under intensive debate. The sensitivity of neutrons to hydrogen makes neutron scattering one of the few techniques that is likely to resolve the structural ambiguities in the H_2S phase diagram. PPU will enable the phase diagram to be explored as a function of pressures up to the 100 GPa range, which is crucial to understanding superconductivity at higher pressures.

Another area of intense study is multiferroicity, whereby ferroelectricity in a material is induced by a noncollinear magnetic structure. The temperature–magnetic field phase diagrams of multiferroic materials are fundamental to understanding the phenomenon, and use of pulsed magnetic fields to 30 tesla or higher in conjunction with neutron diffraction will yield important insights. The diffraction signals from samples subjected to the pulsed fields are collected essentially one pulse at a time, with a duty cycle that requires several minutes to recover. The higher neutron fluxes facilitated by PPU are critical for a more useable signal during the actual measurement time.

Finally, combining polarization analysis with INS enables separation of spin and lattice degrees of freedom and can reveal the individual components of the scattering function

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Quantum Spin Liquids. A quantum spin liquid is a state of matter characterized by the entanglement of particles over distances that are long compared to the atomic scale. Such materials hold promise for future applications in quantum computing and electronics. The sensitivity of neutrons to magnetism was used in an inelastic neutron scattering study to reveal a continuum of magnetic excitations, a key signature of the quantum spin liquid state, in a rare earth–based metal oxide ($YbMgGaO_4$), providing new information on microscopic interactions in this material (Paddison et al. 2017).

$S^{\alpha\beta}(\mathbf{Q},\omega)$, which can provide critical information for a wide range of quantum materials. Such capabilities have been developed on the HYSPEC spectrometer. However, current flux limitations make full extraction of components of $S^{\alpha\beta}(\mathbf{Q},\omega)$ from a variety of quantum materials challenging, and the higher fluxes afforded by PPU are needed to make such measurements routine. This could provide valuable insight into the exotic properties of numerous quantum materials, including multiferroics, superconductors, and topological materials.

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Advanced Alloys. High entropy alloys, a new class of materials with exceptional properties such as ductility, electrical conductivity, oxidation resistance, and high-temperature strength, are currently attracting significant attention. The high penetration and differential sensitivity of neutrons to metals with similar atomic numbers has been used in total scattering experiments to reveal information about a phase transition from an ordered room temperature phase to a high temperature molten phase in a high entropy aluminum alloy. This information will drive further development of this and similar alloys through tuning their compositions (Santodonato et al. 2015).

FUNCTIONAL MATERIALS AND CHEMISTRY

Our technology and economy require increasingly advanced and complex materials, processes, and properties. Advanced materials already perform a myriad of functions that impact our everyday lives, from our food supply, including its safety, to transportation to energy generation and storage to medicine and public health. Development of new materials and advanced manufacturing processes will enable entirely new technologies to emerge. Maintenance and expansion of our nuclear and transportation infrastructures into the 21st century will rely on complex materials, including metal alloys, ceramics, and composite materials, that will be designed and engineered to retain high strength under harsh environmental conditions. Evolving energy demands will be met by developing new materials for storage and conversion with applications in rechargeable batteries, fuel cells, and solar panels (see the appendix). Functional materials are therefore at the heart of the technologies, devices, and societal infrastructure that will define the future US economy and provide solutions to present and future challenges in energy, communications, nanotechnology, security, and transportation.

Some materials, such as the electrodes and electrolytes in batteries or industrial catalysts, are designed to enable chemical reactions. Catalysis is currently a trillion-dollar enterprise, ranging from small-scale applications such as automobile exhausts to industrial-scale petroleum refineries and food processing plants. It is estimated that 90% of all chemically processed products involve catalysis at some stage of manufacturing. However, the performance of materials can also suffer from unwanted chemical reactions such as thermal decomposition or corrosion.

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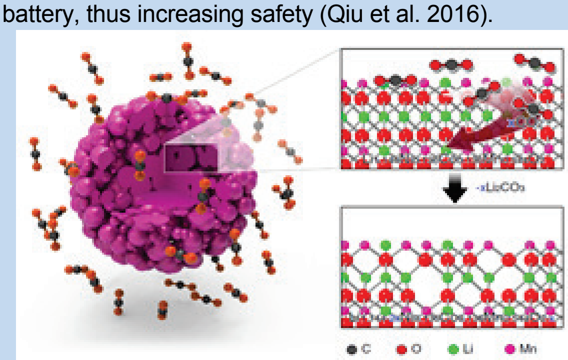
Improved Batteries. The high penetration and high sensitivity of neutrons to small mobile cations such as hydrogen, lithium, and sodium make them a unique probe for obtaining new insights into the operation of batteries. A neutron diffraction study shed light on how changing the oxygen composition of a lithium-rich cathode material could improve battery performance, particularly in high-energy applications such as electric vehicles. By reducing oxygen release during its charging cycle this approach also reduces the flammability of the battery, thus increasing safety (Qiu et al. 2016).



Schematic of gas-solid interface reaction between Li-rich layered oxides and carbon dioxide (Qiu et al. 2016).

PPU will accelerate the development of new materials and new chemical processes in several ways. The increased flux delivered by PPU will make possible measurements on smaller amounts of newly synthesized materials; will enable more rapid and broader access to beam time, thus reducing the cycle time between characterization and design; and will allow access to more extreme environments and better time resolution, making possible routine in operando experiments.

For example, the high penetration and enhanced sensitivity to different elements provided by neutrons is used by researchers to study several types of alloys. PPU will enable measurements to be made with higher temporal, temperature, and/or spatial resolution. This will provide new opportunities for aerospace companies, including GE Global Research, Pratt & Whitney, and Honeywell, and government agencies such as NASA to learn more about the fundamental parameters that drive performance improvement in nickel-based superalloys, titanium alloys, and novel systems such as bulk metallic glass and high-entropy alloys.

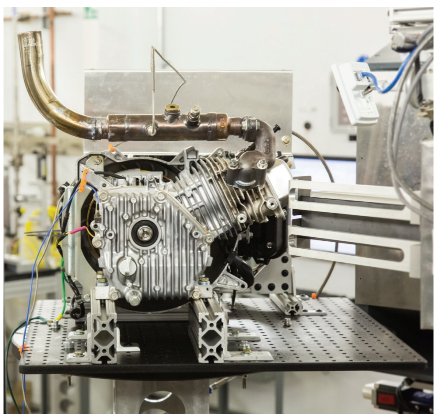


Figure 6. Testing an aluminum-cerium cylinder head using the VULCAN engineering materials diffractometer. (Photo courtesy of Genevieve Martin, ORNL Neutron Sciences Directorate.)

The additional flux provided by PPU will make feasible in situ experiments on materials under complex sample conditions, providing data essential to engineering infrastructure support, including understanding and extending the service life of critical engineering structures. Suspension cables, which are made up of bundles of parallel wire strands, are essential structural components of suspension bridges. Moisture, local defects in the wires, and contaminants can cause corrosion and cracking in the wires. Neutron diffraction on the VULCAN engineering beamline can precisely determine the local strain distribution in wire bundles and has been used to determine the effect of local wire damage on the strain distribution in a bundle, and while additional experiments are currently in progress, in situ experiments are too time-consuming with the current technology to be considered practical. PPU will make them practical and routine.

SNS and HFIR researchers have used several instruments, including the POWGEN and NOMAD diffractometers, the VISION vibrational spectrometer, and the VULCAN engineering instrument (Figure 6), to exploit the penetration and sensitivity of neutrons to small mobile cations such as hydrogen and lithium to obtain new insights into the operation of battery materials and the structure and function of various energy storage devices. While ex situ measurements have become routine, in situ measurements to follow structure and dynamics are still mostly qualitative and often on model systems. With the increase in flux resulting from the PPU project, researchers will be able to quantitatively study the structural and dynamic evolution of electrode and electrolyte materials in real devices.

BIOLOGY

A fundamental goal of modern biology is to develop a predictive understanding of living systems. Gaining this understanding is one of the greatest scientific challenges that we will face over the next few decades and is driven by a desire to improve human health and welfare and our economic and technological competitiveness. This understanding will guide us in protecting and repairing physiological systems, and it will allow us to mimic the architectures and processes of living systems to create new biomaterials such as artificial tissues and bio-inspired technologies including light-harvesting solar cells. It will also provide the information necessary to manipulate microorganisms and their ecosystems to create new biotechnology and biorefinery solutions to emerging energy and environmental challenges.

At the atomic scale, neutron crystallography has been used to provide information about the location of functional hydrogen atoms that are all but invisible to photons. However, it can be difficult to grow crystals of biological molecules large enough to use the available neutron flux on the SNS macromolecular neutron diffractometer, and several user experiments have proven to be impractical because of insufficient crystal size. The increase in neutron flux provided by PPU will enable measurements on smaller crystals, making new experiments possible. For example, DNA polymerases play fundamental roles in maintaining the integrity of the human genome, are linked to cancer, and are potential therapeutic targets. Neutron crystallography is one of the few techniques that could be used to visualize the hydrogen-bonding interactions that are crucial for nucleotide discrimination and polymerase chemical reactions. DNA polymerases have been produced in *Escherichia coli* by fermentation, and relatively large (0.8 mm^3) crystals have been obtained. These crystals diffract neutrons to a resolution of 2.7 Å, which is still not sufficient to resolve the hydrogen atoms in their structures. Increasing neutron flux would help extend this resolution limit and enable visualization of key hydrogen atoms, helping to understand the precise details of how DNA polymerases function. Such insights could guide design of the next generation of cancer drugs. Several other examples at the threshold of feasibility would be enabled by the increased flux resulting from PPU, including greater understanding of (1) the structure, function, and dynamics of cell signaling complexes, which are responsible for transmitting environmental information to cells and mediating cell responses; (2) proteins for developing better antibiotics and drugs against various diseases; and (3) industrial enzymes that have been targeted for improved performance.

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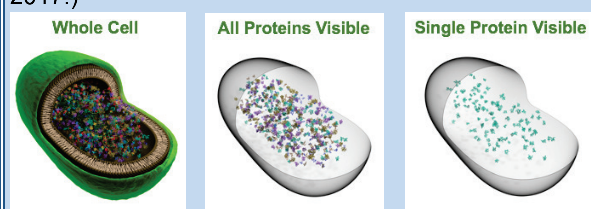
Living Systems. The previously enumerated characteristics of neutrons also make them ideal for studying biological complexes or pathways within living systems. A further advantage of using neutrons is the ability to combine H_2O/D_2O contrast variation with specific deuteration to sequentially highlight different components of these systems (Nickels et al. 2017). For instance, a selectively labeled protein subunit called GroEL was expressed in a culture of living bacterial cells as quasi-elastic neutron scattering revealed important differences in its dynamic behavior within the crowded cytosolic environment compared to in vitro. These subunits form a complex that is essential for protein folding, and defects in their behavior are associated with numerous diseases (Anunciado et al. 2017.)



Selective "highlighting" of parts of biological systems possible with neutrons. (Image courtesy of Robert Standaert, Energy and Environmental Sciences Directorate, Oak Ridge National Laboratory.)

As biologists continue to look beyond static snapshots of individual proteins to problems that require characterizing the assembly of dynamic, flexible functioning complexes and networks, several recent technological advances promise to have a dramatic impact. They include the introduction of direct detector device cameras in biological cryo–electron microscopy and the application of x-ray free-electron lasers in serial femtosecond biological crystallography. These developments allow imaging of individual nanometer-sized particles of complexes and obtaining functionally important dynamic and kinetic information. Researchers are increasingly combining different experimental probes, including photons, electrons, and neutrons, in tandem to develop a more complete understanding of biological systems. Neutrons are an essential part of this new research environment because of the complementary information they provide. The increase in neutron flux from the PPU project will strengthen the impact of neutron scattering in this new multi-technique environment.

For instance, researchers have been monitoring the aggregation process of huntingtin (Figure 7), the culprit in Huntington's disease, by time-resolved SANS on the SNS EQ-SANS instrument to determine the structural consequences that lead to Huntington's disease. The current flux allows a time-resolution of ~15 minutes but with only modest signal-to-noise for the initial time points. Increased neutron flux would enable higher resolution structure determination of the earliest formed aggregate species, which are believed to be the most toxic. Another important biological process that could be studied in greater depth is the formation of fibrin fibers during blood clotting. This dynamic assembly takes place on the time scale of minutes. We know that molecular shear due to the flow of blood plays a role in the formation and orientation of these fibers; however, what exactly happens at the very early stages of this process (the first 30 seconds) is not well understood. While it may be possible to gain insight by following the appearance of weak diffraction peaks from the evolving oriented networks of fibers using time-resolved rheology-SANS, the increase in neutron flux provided by PPU will be needed to bring this into the physiologically relevant realm of the several-second data frame.

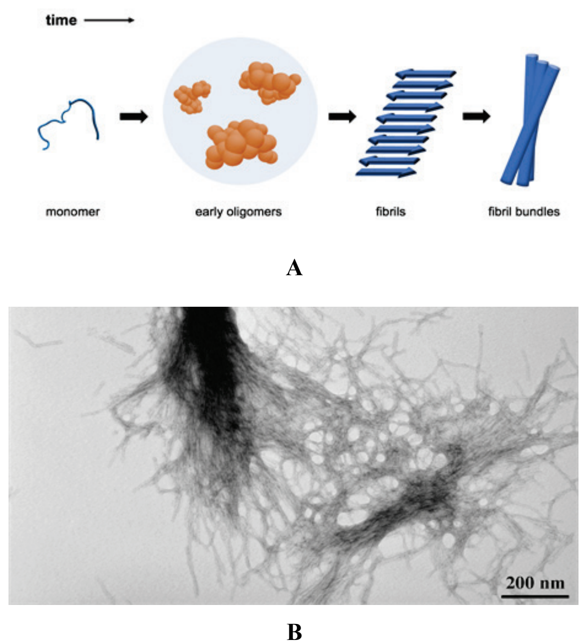


Figure 7. Applications of neutrons in living systems: (A) tracking the formation of fibrils, a hallmark of many diseases such as Huntington's, and (B) investigating the aggregation process of huntingtin, the culprit in Huntington's disease (Perevozchikova et al. 2014). (Images courtesy of Christopher Stanley, Oak Ridge National Laboratory Neutron Sciences Directorate.)

The nondestructive nature of neutron scattering and the ability to use deuteration to highlight specific components or molecules positions neutrons to play a key role in addressing challenges in understanding specific components of complexes or pathways within living systems. Characterizing the various complexes formed at any specific time and cellular location is an emerging grand challenge. These complexes and pathways must be studied directly in the living cell, and neutron scattering provides a potential path forward for doing this. In recent examples, selectively labeled proteins and membrane components have been synthesized in metabolically engineered living deuterated bacterial cells so that their structures and dynamics could be studied. This exciting new approach opens new lines of research on dynamic complexes and pathways using "in cellulo neutron scattering" of biomolecular systems. This may be important as the development of new synthetic biology techniques at the nexus of molecular, cell, and systems biology have positioned biology at the threshold of striking new possibilities that could impact everything from biomedicine to bioproducts to biosecurity. A limitation of studies of these systems is the relatively low signal-to-noise from weakly scattering highlighted components. PPU would help significantly in the future development of this new approach by improving the signal-to-noise.

OUTLOOK

The outlook for neutrons at ORNL is a bright one. PPU will add new scientific capabilities to the SNS FTS that are urgently needed by the research community to address emerging science challenges. These gains will enable new scientific discoveries in the areas of soft materials, quantum materials, functional materials, and biology through experiments that are currently not feasible or routine such as time-resolved in situ measurements, experiments on smaller or less concentrated samples, and experiments under more extreme environmental conditions. An additional benefit of the increased flux delivered by PPU is shorter durations for experiments and therefore an increased number of possible experiments. This in turn will lead to increased researcher access to neutron scattering capabilities and an accelerated pace for scientific discoveries that rely on neutrons to provide unique information about the structure and dynamics of matter. PPU will also provide the capability to produce more proton power to drive a future STS. STS will provide unprecedented access to mesoscale and complex matter, complementing the existing and future capabilities of FTS, with its high peak brightness beams of thermal neutrons at high-repetition rates, and HFIR, with its continuous beams with high time-averaged flux. This three-prong strategy will ensure that US researchers have access to leading-edge neutron scattering capabilities to address critical emerging challenges for the foreseeable future.

REFERENCES

Divina B. Anunciado, Vyncent P. Nyugen, Gregory B. Hurst, Mitchel J. Doktycz, Volker Urban, Paul Langan, Eugene Mamontov, and Hugh O'Neill. "In vivo protein dynamics on the nanometer length scale and nanosecond time scale," *J. Phys. Chem. Lett.* **8**(8), 1899–1904, 2017; doi: 10.1021/acs.jpclett.7b00399.

X. Chelsea Chen, Hee Jeung Oh, Jay F. Yu, Jeffrey K. Yang, Nikos Petzetakis, Anand S. Patel, Steven W. Hetts, and Nitach P. Balsara. "Block copolymer membranes for efficient capture of a chemotherapy drug," *ACS Macro Lett.* **5**(8): 936–941, 2016; doi: 10.1021/acsmacrolett.6b00459.

Ferhat Katmis, Valeria Lauter, Flavio S. Nogueira, Badih A. Assaf, Michelle E. Jamer, Peng Wei, Biswarup Satpati, John W. Freeland, Ilya Eremin, Don Heiman, Pablo Jarillo-Herrero, and Jagadeesh S. Moodera. "A high-temperature ferromagnetic topological insulating phase by proximity coupling," *Nature* **533**, 513–516, 2016; doi: 10.1038/nature17635.

Nature Physics. "The rise of quantum materials," editorial, *Nature Physics* **12**(2), 105, 2016; doi: 10.1038/nphys3668.

Jonathan D. Nickels, Sneha Chatterjee, Christopher B. Stanley, Shuo Qian, Xiaolin Cheng, Dean A. A. Myles, Robert F Standaert, James G. Elkins, and John Katsaras. "The in vivo structure of biological membranes and evidence for lipid domains," *PLoS Biol.* **15**(5), e2002214, 2017; https://doi.org/10.1371/journal.pbio.2002214.

Joseph A. M. Paddison, Marcus Daum, Zhiling Dun, Georg Ehlers, Yaohua Liu, Matthew B. Stone, Haidong Zhou, and Martin Mourigal. "Continuous excitations of the triangular-lattice quantum spin liquid YbMgGaO4," *Nature Physics* **13**, 117–122, 2017; doi: 10.1038/nphys3971.

Tatiana Perevozchikova, Christopher B. Stanley, Helen P. McWilliams-Koeppen, Erica L. Rowe, and Valerie Berthelier. "Investigating the structural impact of the glutamine repeat in Huntingtin assembly," *Biophys. J.* **107**(2), 411–421, 2014; doi: http://dx.doi.org/10.1016/j.bpj.2014.06.002.

Bao Qiu, Minghao Zhang, Lijun Wu, Jun Wang, Yonggao Xia, Danna Qian, Haodong Liu, Sunny Hy, Yan Chen, Ke An, Yimei Zhuk, Zhaoping Liu, and Ying Shirley Meng. "Gas-solid interfacial modification of oxygen activity in layered oxide cathodes for lithium-ion batteries," *Nat. Commun.* **7**, article number 12108, 2016; doi: 10.1038/ncomms12108.

Louis J. Santodonato, Yang Zhang, Mikhail Feygenson, Chad M. Parish, Michael C. Gao, Richard J. K. Weber, Joerg C. Neuefeind, Zhi Tang, and Peter K. Liaw. "Deviation from high-entropy configurations in the atomic distributions of a multi-principal-element alloy," *Nat. Commun.* **6**, article number 5964, 2015, doi: 10.1038/ncomms6964.

Samanvaya Srivastava, Marat Andreev, Adam E. Levi, David J. Goldfeld, Jun Mao, William T. Heller, Vivek M. Prabhu, Juan J. de Pablo, and Matthew V. Tirrell. "Gel phase formation in dilute triblock copolyelectrolyte complexes," *Nat. Commun.* **8**, article number 14131, 2017; doi: 10.1038/ncomms14131.

Thomas Zinn, Lutz Willner, Reidar Lund, Vitaliy Pipich, and Dieter Richter. "Equilibrium exchange kinetics in n-alkyl–PEO polymeric micelles: single exponential relaxation and chain length dependence," *Soft Matter* **8**, 623–626, 2012; doi: 10.1039/C1SM06809A.

APPENDIX. NOTES

1. The scientific productivity of the Spallation Neutron Source (SNS) user program has been increasing each year since it began in 2007. The community of SNS researchers has grown from 165 unique users in FY 2008 to 893 in FY 2016. About 71% of unique users are early-career scientists (under the age of 40), and about 45% are first-time visitors, adding to a user community now numbering more than 3,150 researchers. Users come from academia, industry, government laboratories, and other US Department of Energy (DOE) laboratories. A National School on Neutron and X-ray Scattering, jointly organized by Argonne National Laboratory and Oak Ridge National Laboratory (ORNL), has now trained hundreds of graduate students in the capabilities and use of DOE light and neutron source user facilities, including SNS. The growing SNS research community actively participates in about 15 technical and scientific workshops organized at ORNL each year; its members actively participate in the Neutron Scattering Society of America (NSSA) and provide feedback to SNS staff through the SNS and HFIR User Group (SHUG). More than 200 researchers attended the most recent user group meeting organized by SHUG at ORNL in October 2015 to discuss the proposed Proton Power Upgrade (PPU) to SNS and a future Second Target Station (STS). Growth of the user community has resulted in a large over-demand for access to the neutron scattering facilities at SNS. Requests (proposals) for beam time across SNS instruments greatly exceed availability. The average oversubscription rate is about 300%, but it rises above 400% on some instruments. There is an immediate need to make the neutron scattering capabilities at SNS more accessible to US researchers. PPU is an opportunity to help address this need and is supported by the user community. Together with operational improvements to the SNS source, PPU would increase the number of experiments that could be conducted at SNS, increase the number of users that could gain access to neutron scattering capabilities at SNS, increase the scientific productivity of SNS, and enable critical new experiments that are not possible at present.

2. For the United States to stay competitive and to lead in materials research, it must have the best neutron science capabilities in the world. After a triennial review of SNS and HFIR in August 2012, the US Department of Energy (DOE) Office of Basic Energy Science (BES) requested that Oak Ridge National Laboratory (ORNL) develop a strategic plan to transform the Spallation Neutron Source (SNS) and High Flux Isotope Reactor (HFIR) into centers of scientific excellence and world-leading neutron scattering user facilities. ORNL engaged the research community to participate in developing this plan, which was transmitted to BES in 2013 (Oak Ridge National Laboratory, *Neutron Sciences Strategic Plan 2014*, http://neutrons.ornl.gov/sites/default/files/NScD-Strategic-Plan-2014.pdf). That plan, several DOE- and ORNL-sponsored studies, and workshop reports produced by the research community have all recommended upgrades at SNS that will maintain US leadership in neutron scientific research and innovation. In March 2013, ORNL asked its Neutron Advisory Board (NAB) to provide recommendations for a broad planning process intended to map out future developments at both SNS and HFIR. The NAB recommendations included convening a panel of external experts on future science impacts for neutrons (which took place in June 2013) with the goal to identify areas of science in which neutrons can provide vital information that cannot be obtained by any other technique. Further, NAB recommended organizing a series of focused scientific and technical workshops to produce reports refining those areas of future science priority and to identify new opportunities for neutron scattering in those areas. Four "neutron grand challenges workshops" were convened from 2013 to 2015 to focus on the future of quantum materials (*Quantum Condensed Matter Workshop Report*, December 5–6, 2013, Lawrence Berkeley National Laboratory, Bob Birgeneau, University of California-Berkeley), soft matter (*Grand Challenges in Soft Matter*

Workshop Report, May 17–18, 2014, University of California), biology [*Grand Challenges in Biological Neutron Scattering Report*, January 17–18, 2014, University of California-San Diego (UCSD), Susan Taylor, UCSD, and Heidi Hamm, Vanderbilt University], and materials (*Frontiers in Materials Discovery, Characterization and Application Workshop Report*, August 2–3, 2014, Schaumburg, Illinois, George Crabtree, University of Chicago and Argonne National Laboratory, and John Parise, Stony Brook University and Joint Photon Sciences Institute). These areas encompass and directly map to the transformative opportunities identified in the BES Grand Challenges update (*Challenges at the Frontiers of Matter and Energy: Transformative Opportunities for Discovery Science*, https://science.energy.gov/~/media/bes/besac/pdf/Reports/Challenges_at_the_Frontiers_of_Matter_and_Energy_rpt.pdf). Quantum materials maps most directly to harnessing coherence in light and matter, while soft matter and biology are aligned primarily with mastering hierarchical architectures and beyond-equilibrium matter, and the materials workshop explored many of the topics in beyond ideal materials and systems (e.g., understanding the critical roles of heterogeneity, interfaces, and disorder). The scientists from all four workshops noted that in each of the areas covered, neutrons would play a unique and powerful role in understanding structure and dynamics in materials required to develop future technologies. Participants concluded that world-leading neutron facilities such as an upgraded SNS will be needed to complement other forefront advanced research user facilities in addressing the mission of DOE. PPU will enable currently unfeasible experiments to be conducted at SNS in science priority areas. Participants also identified emerging grand challenges that will require the enhanced capabilities of an STS to provide gains in performance of 2 orders of magnitude and beyond in areas of science that require high peak brightness of cold neutrons at low-repetition rates. Overall, STS will provide unprecedented access to mesoscale and complex matter, complementing the existing and future capabilities of FTS, with its high peak brightness beams of thermal neutrons at high-repetition rates, and HFIR, with its continuous beams with high time-averaged flux.