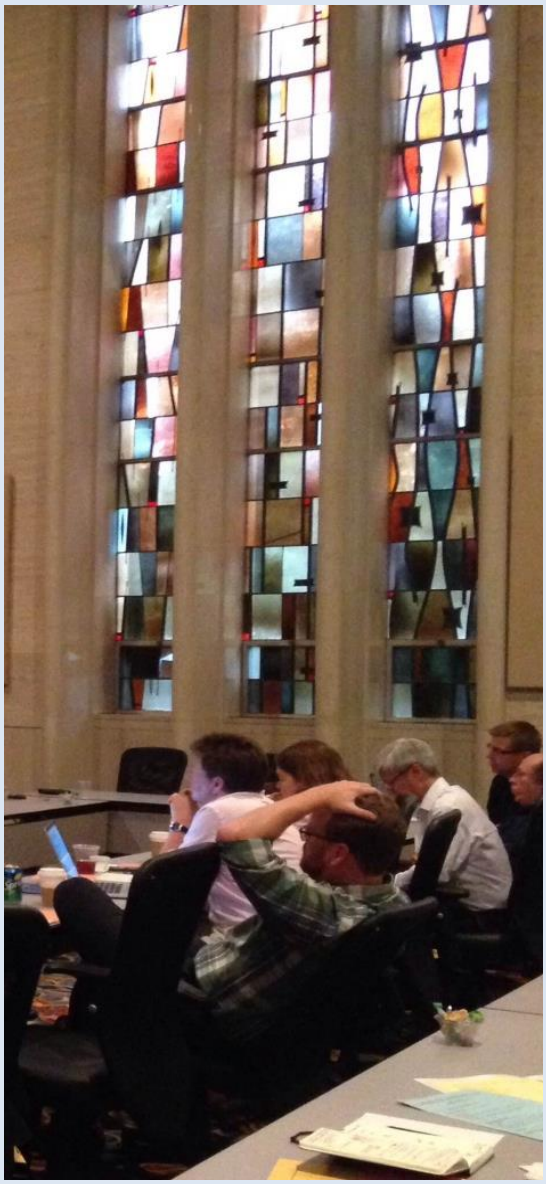
Neutron Measurements for Materials Design & Characterization

Workshop Report

August 21-22, 2014
Bolger Center
Potomac, MD



NIST Center for Neutron Scattering Workshop on
Neutron Measurements for Materials Design and Characterization
Aug 21-22, 2014 at the Bolger Center, Potomac, MD

Co-Chairs: Rob Briber, University of Maryland
Collin Broholm, Johns Hopkins University

Thurs August 21 Morning
Plenary talks (30 min)
8:30 – 10:40am
Rob Dimeo, NCNR Director – "NCNR Facility Overview"
Collin Broholm, Johns Hopkins University –"Impacts of Neutron Scattering on Hard Condensed Matter Physics"
Norm Wagner, University of Delaware – "Probing the Dynamics of Soft Matter with Neutrons"
Mike Toney, SSRL - "Neutrons and X-rays in Sustainable Energy Materials"

10:40-11:00am Break

Thurs August 21 Morning (cont.)
Technique Development talks (15-20 min each)
11am – 12:00pm
Chuck Mielke, Los Alamos National Laboratory – "Pulsed High magnetic Fields for scattering sciences"
Dan Hussey, NIST – "Ballistic Guides and Imaging Forming Mirrors: How to simultaneously improve resolution and intensity"
Chuck Majkrzak, NIST – "Polychromatic Beam Neutron Reflectometer Employing Energy Dispersive Detectors"

Thurs August 21
12:00-1:00pm Lunch in the Osgood Dining Room

Thurs August 21 Afternoon and Evening
1:00-5:30pm Breakout sessions
6:00-7:30 pm Working dinner for Subcommittees in the Osgood Dining Room at the Bolger Center
8:00pm – 10pm Breakout sessions (continued)

Fri August 22 Morning
8:30am – 12:00pm Breakout sessions

Fri August 22

12:00-1:00pm Lunch in the Osgood Dining Room

<u>Fri August 22 Afternoon</u>
1:00-3:00pm
Presentation of Reports from Subcommittees

Breakout Groups

Session Title	Chair
Innovative Sample Environments	Roger Pynn, Indiana U.
Polymers and Colloids: Structure	Mark Foster, U. of Akron
Biological Systems: Structure	John Tainer, LBNL
Soft Matter Dynamics	Ramanan Krishnamoorti, U. of Houston
Energy Conversion and Storage Materials	Mike Toney, SLAC
Condensed Matter Physics	Stephan Rosenkranz, ANL
Materials Chemistry	Angus Wilkinson, GaTech
Engineering Materials	Don Brown, LANL
Nanostructured Hard Materials	Steve May, Drexel U.

Neutron Measurements for Materials Design & Characterization Workshop
August 21-22, 2014

Executive Summary

The success of America's industrial and academic institutions and America's role as an economic leader in the world is built upon innovation and entrepreneurship in fields of science and engineering. Maintaining this leadership requires pushing the boundaries of discovery and the constant creation of new knowledge to provide a pathway for the development of new technology. Neutron scattering is a key measurement technique for elucidating a broad range of physical phenomena ranging from the atomic structure of new materials, the complex interplay of electronic and magnetic behavior, the interactions and flow of colloids and macromolecules and the dynamics and superstructure of complex biomolecule assemblies. In the era of big data and combinatorial materials synthesis, neutron scattering is a leading tool for guiding the design, discovery and characterization new materials. The NIST Center for Neutron Research (NCNR) has been at the forefront of neutron science for the past 20+ years and with the construction of a new instrument guide hall, the installation of five new neutron guides , a new fundamental physics station, improvements in current instruments, novel detector technologies, two innovative new instruments (VSANS, CANDoR, and an imaging station with a neutron microscope) under development, and planning for a new D_2 cold source, the NCNR is poised to remain a leader in neutron research in the U.S. and the world for many years to come.

With the new guide hall there are additional opportunities to strengthen the science in both hard and soft matter.

Soft Matter
Neutron scattering has provided fundamental insight into the structure and dynamics of soft matter for more than 40 years through deuterium labeling and by exploiting the large incoherent cross-section of hydrogen. With advances in optics and instrumentation there are a wealth of opportunities for new understanding in colloid and polymer science and in biology using neutron scattering. Recent advances in synthesis of designer colloid systems allow sophisticated control of particle size, shape and surface chemistry that provide a large design space for self-assembled structures. Many of the most important applications of polymers involve multicomponent systems with end-use properties that are optimized by kinetically trapping the nanoscale structure. Neutron scattering can provide both an understanding of the structure and information on the kinetics of structural evolution and local dynamics within the morphology. In contrast to bulk systems, polymer surfaces and interfaces hold the key to our understanding of some physical processes. For example the location of just a few layers of water molecules can be tracked using neutron reflectometry during the initiation of corrosion or the operation of proton exchange membrane fuel cells.

The wealth of information on biomolecular structure provided by X-ray diffraction, NMR and cryo-TEM over the past 50 years has helped build an understanding of biological function on the molecular level. It is now apparent that there are many supramolecular assemblies of proteins with DNA, RNA, lipids and other biomolecules that play a critical role in cellular function. Neutron scattering can provide powerful insights into the structure and local dynamics of these assemblies through the use of selective deuteration to highlight (or contrast match) parts of the

system. By leveraging the NIST/IBBR Biomolecular Labeling Laboratory (BL^2) specific labeling of proteins can be done to examine the dynamics of disordered proteins, protein aggregation and elastic properties of lipid arrays.

The penetrating power of neutrons makes many types of *in-situ* experiments feasible that are much more difficult using other techniques such as X-ray scattering. For example many of the most interesting soft and biological materials are strongly influenced by external fields (electric, shear, pressure) and neutron scattering can provide fundamental insight to the behavior of soft materials under these conditions. Similarly neutron scattering could provide real time information on the local diffusion and mixing process occurring during additive manufacturing through interfacing a 3D printing system on a small angle scattering beamline.

Hard Matter

Neutron scattering has had tremendous impact on our understanding of hard matter ranging from fundamental condensed matter physics to nanomaterials to materials for energy conversion and storage. The ability to probe structure and dynamics over a broad range of length and time scales is indispensable in areas that include hard and soft magnets, quantum correlated materials, high-T_c superconductivity, multiferroic, ferroelectric, and thermoelectric materials. Because their nuclear and magnetic interactions with materials are of similar strength and can be distinguished in polarized beam experiments, neutrons are essential to disentangle the complex interplay of charge, spin, lattice and orbital degrees of freedom that can produce unique physical properties. It is also important that neutron scattering can be applied to bulk and nanostructured solids under a wide range of thermodynamic conditions that mimic applications or expose novel properties.

While quantum coherence beyond the atomic scale is best known from superconductivity, it can also occur in magnetic materials when competing interactions frustrate the development of conventional magnetic ordering. One of the largest focus topics at the annual APS meeting, the field of frustrated magnetism is uncovering fundamentally new forms of quantum correlated matter with potential impacts in quantum information processing using neutrons as the preeminent experimental tool.

Neutron scattering is also essential to understand and control materials in the technologically important area of spintronics where functionality is associated with the spin of the electron rather than its charge. Thin film neutron diffraction was thus cited by the 2007 recipients of the Nobel Prize in Physics as critical to understanding the phenomenon of Giant Magneto Resistance, which is the key technology in more than 5 billion read heads for gigabyte hard disk drives shipped since 2007.

Materials Chemistry

Technological breakthroughs are often derived from discoveries of new materials. It is through rational design and discovery of new materials that the US can maintain technological leadership in the world. This includes high throughput synthesis and screening of materials such as metal oxide framework and zeolites, piezoelectrics, superconductors, ionic conductors, *etc*. as outlined in the Materials Genome Initiative. These materials often combine many different atoms in large unit cells with subtle distortions of atomic positions and bond lengths that give rise to interesting and useful properties.

With increased interest in materials for energy conversion and storage there is a major opportunity for neutron scattering to provide non-destructive characterization of complex systems such as Li-ion batteries, proton exchange fuel cells and carbon sequestration materials *in-operando*. Neutron scattering can play a complimentary role to electron microscopy and X-ray scattering in charactering thin film organic and heterojunction photovoltaics, supercapacitors and solid electrolyte batteries.

Nanostructured materials such as quantum dots, nano- rods and particles, topological insulators, and catalysts for use in next generation magnetic storage media, quantum computers, nano-medicine and advanced chemical synthesis represent an wealth of scientific opportunities where neutron scattering will play an important role. Many of these systems will require the state of the art neutron instrumentation associated with the new NCNR guide hall and future D_2 cold source to provide the range of brightness, Q-range, and energy transfer to characterize effectively.

Engineering

Neutron diffraction measurements of residual stress in solids has proven to be a very valuable technique for examining bulk materials and manufactured objects both prior to service and post-service for failure analysis. Due to their penetrating power, neutrons are particularly powerful for analyzing residual stress in dense materials and for measurements on large specimens at high diffraction angles. Additionally, innovative sample environments for realistic, multi-axial materials deformation will enable the measurement of mechanical properties of engineering materials in ways not available to other methods. This combined with the ability to exploit neutron cross-sections to examine structures associated with light elements (hydrogen) and differentiate elements such as Co and Fe makes neutron diffraction a unique tool for understanding engineering materials.

Instrumentation and Facilities

With the development of VSANS and CANDoR, the promise of a new D_2 cold source, improvements in many other instruments and new detector technologies, the NCNR will remain one of a small number of leading neutron scattering facilities in the world. There are still open end positions available (one served with thermal neutrons and one with cold neutrons). These positions are quite valuable and new instruments should be built at each after a careful evaluation of proposed instrumentation is carried out. Some proposed instruments include:

High data rate materials diffractometer – this is particularly relevant given the emphasis on materials discovery and high throughput characterization techniques and the Materials Genome Initiative. Such an instrument could play a role across many fields of science and technology, for example when a new material is identified with unexpected properties (such as a new high Tc superconductor), for screening large numbers of newly synthesized compounds or for following the kinetics of temperature or field driven phase transformations or synthesis of novel materials in real time.

High wave-vector resolution multiplexing spectrometer – A new concept for high efficiency neutron spectroscopy at a reactor-based source is proposed where the detection system *simultaneously* covers a wide range of scattering angles and a full spectrum of neutron

energies. Such an instrument would retain the high momentum resolution of a conventional cold triple axis spectrometer such as the SPINS instrument while gaining at least two orders of magnitude in data rate. The proposed instrument would constitute a powerful new tool for the elucidation of collective phenomena at the mesoscale in hard condensed-matter physics.

Biological SANS instrument – there has been a resurgence of interest in small angle scattering in the biological research community over the past 10 years with a dramatic increase in small angle X-ray publications and a similar increase (but at a lower level) in SANS publications. The current NCNR SANS instruments are oversubscribed and are likely to remain so for the foreseeable future. A biological SANS instrument should be optimized for beam brightness at the sample position to allow for smaller sample volumes and lower concentrations ($\leq$100 µl or less with $\leq$1% sample concentration).

Upgraded NSE instrument – neutron spin echo which is essential for elucidating details of the motions of materials provides the highest energy resolution available with neutron scattering. The recent upgrades to this instrument have improved the performance by a factor of 3. Even larger gains could be realized by improvements in the magnetic field correction elements, replacing the resistive main coils with superconducting coils and by providing better focusing at the sample position through the use of Wolter optics or other focusing schemes.

There should be a concerted effort to develop new neutron optics (focusing optics and Wolter optics) to provide high flux and high brightness neutron beams for all instruments. In many cases increasing the scientific impact and user base for a technique is limited by the flux and brightness of the incident beam. For example, neutron spectroscopy of soft materials could make many substantial new scientific contributions with improvements in optics to allow for sample volumes on the order of 100 µl. The implementation of brighter neutron beams will allow for *in-situ* experiments, irreversible time-resolved kinetic measurements, smaller and more compact sample environments and the opportunity to examine a broad range of novel devices (batteries, microfluidics, chromatography systems, *etc*.) while in operation in the neutron beam.

NIST has played a leading role in the development of neutron polarization techniques through the use of spin filters based on polarized ^{3}He. This is having a major scientific impact in hard condensed matter experiments. Thus these efforts should be expanded to more instruments. Moreover NIST should continue to work at improving all aspects of the performance of these devices with a particular goal of reaching at least 90% polarization of the ^{3}He nuclei in the filters.

The large penetration depth of neutrons allows for a range of sample environments including shear, pressure, electric and magnetic fields. Although these areas have been explored, there is a need to expand the availability of such sample environments to all instruments and to engage the scientific community in identifying and developing new systems that extend the range of the applied fields.

In addition to the capabilities at the neutron spectrometer in terms of sample volumes, sample environments, *etc*., there needs to be a concomitant level of support for the users in terms of local lab facilities, software and scientific expertise at the facility. What often differentiates one user

facility from another is the quality of the support infrastructure in terms of lab space for sample preparation, ancillary characterization tools, high quality data collection, visualization, and analysis software and staff expertise in the relevant science, modeling, and analysis. Having data collection and visualization software that can be learned in the time frame of 1-2 hours benefits both external users and instrument scientists. Too often improvements in data collection, visualization, and analysis software take the lowest priority and results in frustration among users and reduced scientific productivity.

Access to neutron scattering beamtime is a valuable national resource and throughout the world most neutron facilities report that the majority of their instruments are oversubscribed by a factor of 1.5-3x. With the expansion of detector arrays, data sets have become increasingly large and complex, particularly when combined with time resolved measurements as a function of temperature and/or external field. This makes it difficult for the original experimentalists to extract and publish all scientific results supported by the data in a timely fashion. NIST already makes all raw neutron data available to the general scientific community. Thought should be placed on improving both the searchability of these datasets and the provision of relevant supplementary information to fully define the experiment. In this way, NIST could play a key role in facilitating expedient analysis of neutron scattering data by engaging a broader array of scientists.

Through the implementation of the on-going and planned upgrades to the facility and new instrumentation and by following the science outlined by the participants at this workshop, the NCNR will remain a world leader in neutron science for the next decade and beyond.

Innovative Sample Environments Working Group

While many individual recommendations for new sample environments are included in sections on specific scientific drivers for facility development, it is useful to examine the provision of sample environments for users in a more strategic manner. Several overall directions for development emerged from our discussions. In summary, we suggest that pursuing the following overall themes provides an avenue for continued improvement of the scientific, technical and societal impact of research performed by users of the NCNR:

1. Probing non-equilibrium phenomena
2. Using neutrons to study operating devices
3. Designing sample environments to increase the efficiency of neutron measurements
4. Engaging a larger scientific community in the development, design and construction of sample environment equipment for the NCNR
5. Extending the dynamic range of the thermodynamic parameters provided
6. Deploying neutrons and other measurement techniques simultaneously

In the following paragraphs we present an incomplete set of examples that support these strategic directions.

1. Probing non-equilibrium phenomena

With the increase in effective neutron flux incident on samples as well as event-mode data collection, it is becoming possible to probe the kinetics of a diverse array of systems on relevant time scales. This is particularly true of soft matter where the characteristic time and length scales of interest are intrinsically coupled through the fluctuation-dissipation theorem, with relatively small energy scales, set either by the Brownian temperature or by the imposed shear stress. Structural evolution can vary over many orders of magnitude from segment level orientation to molecular level deformation to supramolecular phenomena such as flow-induced phase transformations and demixing. Corresponding timescales may vary from microseconds to seconds. Detection systems currently under development such as the VSANS instrumentation afford the desired capabilities such as high fluxes over a wide range of q vector and there is an imminent need to develop appropriate devices and fluid systems that can take advantage of this instrumentation. Other features of strong interest include the ability to perform faster time-resolved measurements that synchronize mechanical, thermal and electrical data with the neutron data (e.g. using stroboscopic techniques, beam-chopping, etc.) as well as direct imaging of structure evolution on a wide range of length scales e.g. using coupled X-Ray/Neutron imaging).

Using TISANE instrumentation already installed on one of the SANS instruments at the NCNR there is currently an opportunity to explore the behavior of systems under the influence of periodic fields (electric, magnetic, light, stress etc) with various waveforms and with sub-millisecond time resolution. Examples of fields where this technique could be applied include: the motion of magnetic vortices in superconductors; electro-

rheological fluids; correlating structure with dielectric or magnetic ac susceptibility; and tracking the development of distributed damage in materials due to fatigue.

2 Using neutrons to study systems in situ and in operando

There is a critical and unmet need for measurements of complex fluids in extreme environments that mimic industrial processes. Currently it is difficult or impossible to directly characterize structure-property relationships in fluids where micro- and nano-scale confinement can lead to exceptionally high shear rates, capillary condensation, surface tension effects, and structures that are dominated by surface driven phenomenon (local ordering, adsorption/desorption, aggregation). Unfortunately, despite the measurement difficulties, these conditions are commonly encountered in many industrial applications.

High shear rate flow of fluids (millions of strain units/s) occurs in many industrial and commercial processes including lubrication, mixing, spraying and injection. Some challenges that can arise in these extreme contexts include clogging or degradation of the fluid. Similarly, there are concerns that high shear rate delivery of protein therapeutics could affect structure and drive aggregation with toxic and potentially fatal effects. From these contexts, biopharmaceutical and specialty chemical companies have expressed a need for simultaneous measurements of rheology and structure at shear rates exceeding 10^6 s^{-1}. Unfortunately, there are no viscometers built for neutron scattering that can exceed shear rates of 10^4 s^{-1}. Additionally, we suggest the development of devices for non-shear and mixed flows that are commonly encountered in industrial settings.

Although the new sample environments will probably be designed initially for SANS measurements, devices should be designed, where possible, to be compatible with inelastic scattering techniques to measure the dynamics of flowing complex fluids.

With the strong emergence of natural gas extraction and enhanced oil recovery, there has been a dramatic increase in large-scale material development to enable this industry. Often the materials used in these technologies are complex fluids composed mainly of soft matter materials; polymers, surfactants and solvents that are designed to perform under rapidly changing conditions under high temperatures (~ 300°C), pressures (~ 100MPa), stain rates and through porous media. In response to this, there is a strong industrial and academic desire to understand the behavior and structure of complex fluids under these conditions. For instance, a structured surfactant system which contains a favorable viscoelastic profile can easily undergo flow instabilities and structural rearrangement under these rigorous conditions, which could have a large effect on their performance. Although current characterization techniques, both neutron and otherwise, can robustly measure rheology and structure of complex fluids, there is currently no method that can measure both simultaneously at temperatures, pressures, and physical environments (i.e. porous media) relevant to these applications.

Over the past five years great strides have been made in developing rheo-SANS sample environments that enable interrogation of the non-equilibrium structures of soft

and deformable matter under flowing conditions. As a natural first step, the primary focus has been on steady simple shear flows with imaging in the 1-3 (flow vorticity) and, more recently, the 1-2 (flow/shear) plane. The important next step is to extend these capabilities to more industrially relevant flows that characterize real processing flow geometries of interest to US industry; these include spatially inhomogeneous shearing flows (e.g. pressure-driven flow in pipes and ducts), as well as mixed flows that combine distinct regions dominated by extensional and shearing kinematics (e.g. mixing flows and converging nozzles, spin lines etc.) Such flows are ubiquitous in the process industries and understanding the coupling between rheological response and morphological development is essential in developing a fundamental understanding of the soft solids and complex liquid systems that form the basis of almost all advanced consumer products, food stuffs and polymeric engineered materials. Sample environments that address this problem should be able to vary the geometry, length scale, material of construction and dynamical characteristics of the flow geometry under examination. These needs can be addressed at the *millifluidic* level by introducing and optimizing key enabling technologies such as 3D printing and other rapid prototyping tools (e.g. water-jet or wire-EDM cutting of masks and templates) as well as well-established soft microfluidic fabrication approaches (e.g. SU-8 masks, metallization etc.) for making a wide range of model flow geometries.

3 Designing more efficient sample environments

There are several senses in which the design, construction and operation of sample environment equipment could be made more efficient. These range from collaboration between facilities or with users on the design of new environments, through the use of new materials and manufacturing techniques such as additive manufacturing, to combining sample environments with focusing optics to improve the signal-to-background ratio of experiments.

The group heard that European neutron facilities will prepare a common standard protocol for interfacing sample environment equipment to instrument control software. This standard will be defined in a way that it is compatible with a broad variety of soft- and hard-ware operated at the different large scale facilities (LSF). Special emphasis will be given to compatibility with laboratory-based equipment, processes and protocols used in industrial R&D and at different LSFs (including synchrotron radiation facilities). The protocol will also include real-time acquisition systems for engineering and applied sciences of particular relevance for future industry-based users. In a few years, implementations of the Sample Environment Communication Standard Protocol (SECoSP) will be tested at different facilities and provided to interested industrial partners for implementation in their commercial scientific tools (e.g. Cryogenic, Oxford Instruments). We learned that an international society is being set up and will soon organize a first school for training technicians, engineers and scientists to the operation of standard equipment. There is an opportunity for the NCNR to participate in this

activity at this formative stage and to both learn and influence future outcomes.

In the context of SECoSP all sample environment related metadata will be made available in a standard form and will be appended to the data collected during an experiment.

The adoption of these standards at NCNR would greatly facilitate the installation and use of new equipment developed by other facilities, industrial partners or even internally.

To improve the signal-to-background ratio of standard equipment like cryostats, furnaces, humidity chambers, pressure cells, etc. novel materials can be used. For example, the screens of the cryostats and cryofurnaces are made from soft aluminium but can be replaced with much thinner windows made from harder aluminium alloys. The amount of material in the beam can be decreased by a factor 5 without impacting the transparency. Pressure cells made in CuBe can also be replaced with hybrid cells made from different materials that lead to better performances without impacting the transparency (multilayer technique).

Extreme conditions being only applicable over very small volumes, we also suggest the introduction of focusing optics. To be efficient, these optics will have to be installed outside and inside the sample environment equipment. Gain factors of more than 2 are expected as revealed by studies performed by PSI (prototype) and ESS/ILL (simulations using genetic algorithms). Because sample environment devices aligned with outside optics cannot be tilted, we also suggest the implementation of goniometers for orienting crystals inside cryostats and cryomagnets. Compact goniometers are already available elsewhere.

4. Engaging a larger scientific community in the development, design and construction of sample environment equipment for the NCNR

While the experience with engaging the user community in the development of neutron-related sample environments has been mixed, the group believes that exploration of this avenue should continue. Recent experience in Europe indicates that with sufficient coordination between users and the facility, sample environment equipment can be developed effectively. With such coordination, equipment that is usable by the broader scientific community and not just by a few experts can be built and integrated with various spectrometers. In this context, the experience of the NHMFL may provide a useful model.

5 Extending the dynamic range of the thermodynamic parameters provided

There will continue to be a need in various communities for an increase in the range of the thermodynamic variables (such as temperature, magnetic field, pressure etc) that

can be applied to samples. For example, the development of high magnetic fields coupled to neutron beams is a potential area of growth signaled by the National Academy of Sciences and driven by new phenomena. The use of high magnetic fields in condensed matter physics is ubiquitous as a probe of the relevant energy scale between the constituent electrons within a system. The binding of electrons in the Cooper pairs that are involved in high temperature superconductivity, for example, is much larger than the binding of electrons in an average metal, and often requires large magnetic fields to probe (ie. greater than 15 T). Key advances in highly correlated electron systems such as heavy fermion metals, highly frustrated magnets, and novel superconductors have been made by other thermodynamic probes in high magnetic fields at laboratories such as the NHMFL. This has led to the elucidation of the high field magnetic phase diagrams of these systems, the unraveling of the mechanisms behind the coupling of magnetic and electric fields in multiferroics, or the development of solid state qbits for quantum computing. The NCNR can make a large impact in this field, especially with the extra quiet guide hall where instruments can be isolated away from such experiments that might disrupt techniques such as spin echo. Since neutron scattering is the premiere method for visualizing magnetic structures and excitations, it is natural to extend these techniques to fields greater than 20 T, where many highly correlated phenomena begin to appear, such as the emergence of quantum criticality. The development of such fields, via pulsed or continuous sources, or through monochromatic or polychromatic beams, would have to be explored in detail involving collaborations from multiple sources.

The application of high pressure to condensed matter systems is a complementary method to explore the phase diagram of complex materials. Pressure and magnetic fields are both methods, for example, to explore quantum critical points in materials or to suppress magnetic ordering or superconductivity. The development of new high pressure cells that can be used in tandem with low temperature cryostats is an obvious area to explore for future sample environments. High pressure research is also related to developments in materials chemistry such as hydrogen storage compounds, the phase diagrams of van der Waals solids such as ice, and exotic phases of hydrogen. Geologists have an interest in chemical phases under pressure similar to what is found deep within planetary structures. The capability to measure high pressure low/high temperature phases (ie. below liquid helium temperatures, at temperatures of the Earth mantle) via neutron scattering is largely a niche area in the United States that needs further development. Considerable development of neutron optics would have to be undertaken as well to accommodate small sample sizes involved with such sample environments.

6 Simultaneous measurements using other techniques

Finally, there is a need for many of the techniques to be used in tandem (i.e. some combination of high field, low temperature, or high pressure experiments). These

would require considerable effort and innovation from the sample environment team to adapt such probes on different instruments. The phase diagrams that could be reached by the combination of these techniques would be a unique capability for the NCNR and greatly expand the science that can be accessed with neutron spectroscopy.

It is rarely true that neutrons provide the only information needed to understand a particular phenomenon. Usually other measurements, such as measurements of various susceptibilities, need to be correlated with the structural and dynamical information provided by neutrons. However, as increasingly complex phenomena are studied with neutrons, it is becoming more important that these complementary measurements be made simultaneously. For example, in the study of soft materials a key requirement is to simultaneously probe *in situ* not only the microstructural response of material (using the appropriate neutron scattering technique) but also the rheological, thermal, mechanical or electrical response of the same material system at the same time. This is essential to provide a direct unambiguous correlation between the measured structural and rheological response which enables fundamental morphological understanding to be translated back to home laboratories – which typically have access to rheometry, calorimetry, and other diagnostic techniques that can be used to explore formulational variations (e.g. changes in temperature, concentration etc.) but no ready access to neutrons. Designing sample environments that enable such thermal and mechanical tests to be performed *in neutro* would enable the development of *neutro-mechanical* test protocols that mirror the rapid explosion of electro-optical and thermo-mechanical test systems over the past few decades. Specific examples might include the ability to simultaneously measure the shear and normal stress growth and microstructural evolution during the start up of steady shear flow of a complex fluid, or the ability to monitor the simultaneous evolution in percolated structures of fractal carbon black nanoclusters with EIS measurements of the fluid conductivity during charging and discharging of prototype flow battery materials for grid-level energy storage.

POLYMERS AND COLLOIDS WORKING GROUP (STRUCTURE)

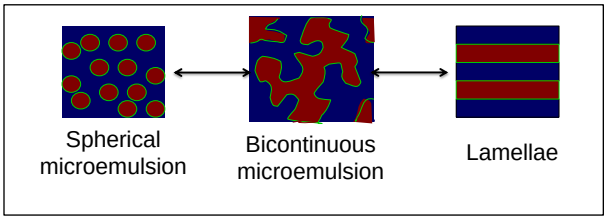
Participants: Mark D. Foster (Chair), Dan Blair, Robert Briber (Workshop Chair), Boualem Hammouda, Matthew Helgeson, Yun Liu, Srinivasa Raghavan, Megan Robertson, Sushil Satija, Rafael Verduzco,

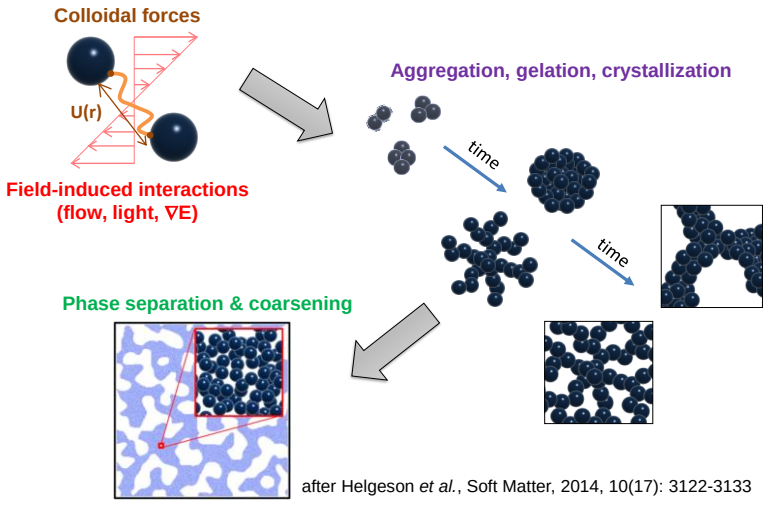
Scientific opportunities and Impact:

We highlight five topic areas of scientific opportunity in which the timely exploitation of developments in neutron instrumentation will have significant scientific and technological impact. These five areas are-

i.) kinetics of multiscale structure transformation in blends, ordered polymeric and supramolecular materials and colloidal systems,
ii.) field driven structure formation,
iii.) bio systems and biomimetics/bio-inspired systems,
iv.) structure at interfaces of polymers and colloidal materials, particularly in the first few molecular layers,and
v.) polymers and colloids under confinement

Polymer blends provide new and valuable properties through the combination of polymers, with each providing favorable attributes. Most polymers in blends are not miscible when mixed. Thus most combinations result in inhomogeneous materials, with phase separation sometimes occurring on length scales that can be probed well with neutron scattering techniques. In many cases, polymeric surfactants, known as compatibilizers, are required to manipulate the blend morphology to provide optimized mechanical and thermal properties. Though many studies have addressed the equilibrium phase behavior of compatibilized polymer blends, little is known about the mechanisms of evolution from one equilibrium phase to another. In addition, the real-time response of the blend morphology (either with or without the addition of a surfactant) to various types of processing (i.e. heating or subjecting to elevated pressure, shear or other deformation forces) cannot be probed with existing small-angle neutron scattering methods, which typically require collection times on the order of minutes to an hour for a single data set. Data collection times on the order of seconds or smaller could probe in-situ the response of such morphologies to external stimuli. Analysis of the experimental results would be exploited to develop precise control of the evolution of the material structure during blend processing. This control would impact the design of lightweight parts from polymeric blends



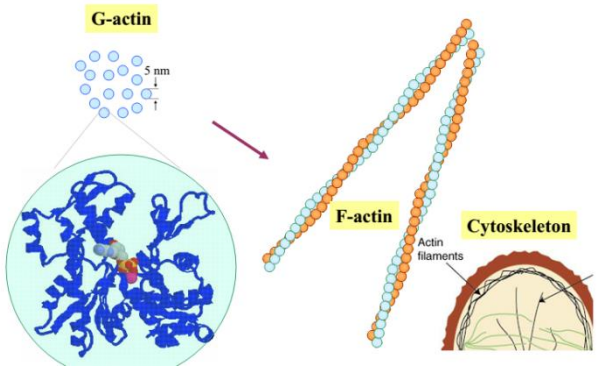


Examples of multi-scale kinetic processes in colloids.

and processes for their manufacture, with applications in energy, human health, consumer goods, and structural composites.

New capabilities to synthesize designer colloids with sophisticated control over particle size, shape and surface chemistry have opened up a vast design space of building blocks, but with few design rules for achieving specific assembled structures. A longstanding and unresolved obstacle is the lack of understanding of the kinetics of colloidal assembly at the particle scale and also the aging and coarsening that occur at larger length scales. Of particular need is an understanding of the evolution of non-equilibrium, kinetically arrested states the aging of which depends critically on collective and heterogeneous structural rearrangements. Understanding these phenomena would lead to improved engineering of particle morphology and their interactions in order to assemble with high specificity desired structures and targeted properties. These capabilities would open up a number of new technologies and lead to improvements in self-assembled photonic materials, advanced structural materials and composites, and more rational formulation of consumer products and pharmaceuticals.

A second area of inquiry closely related to the first is that of field driven structure formation. Polymer blend, block copolymer, nanoparticle composite and colloidal structures may be strongly impacted by the imposition of fields including flow fields, thermal gradients, chemical gradients, and electric and magnetic fields. Advances are needed in the understanding of nonequilibrium structures formed under the influence of these fields. Those advances will increase the range of materials that can be produced from a palette of components and impact the efficiency of manufacturing processes.



Bioinspired or biomimetic materials range from fluids such as blood and milk to soft solids like individual tissues and hard solids like bone or mollusks' shells. They constitute a third area of opportunity. A critical feature of these systems is that they are structured from the bottom-up across a wide range of length scales extending from the nano- to the milli- or centi-meter scale. In the structure of the actin cytoskeleton present in eukaryotic cells (Figure 1) the nanoscale protein G-actin assembles into nanoscale helical filaments (F-actin); the filaments then bundle into thicker fibers; the fibers become entangled and cross-linked by other proteins to form a three-dimensional network (gel). Researchers are seeking to create synthetic materials exhibiting the same hierarchical structure. Custom-synthesized peptides have been found to form in water hierarchical gels much like those of actin. In a similar vein, researchers are seeking to create "nano-cellulose", which retains the hierarchical structure of cellulose, and acts as an excellent reinforcing agent for many plastics. Neutron scattering studies revealing the hierarchical structures over multiple decades in length scale will lead to new reinforcing agents like nanocellulose and to improvements in human health, e.g. through the development of artificial organs and in the treatment of diseases.

Elucidating the structure at the interfaces of polymers and colloidal materials, particularly in the first few molecular layers, is our fourth area of opportunity. The structure of water at these interfaces is a subject of intense interest in subfields such as corrosion, interactions of polymers and colloids in biological milieus, and the effect of water content on polymer containing devices such as solar conversion systems. Neutron reflectometry and grazing incidence scattering measurements that can resolve structure on this very small length scale will impact transportation infrastructure (bridges, roads, vehicles), energy production and distribution,

corrosion prevention and mitigation (which is a $20B+ problem in DoD alone), the reduction in biofouling in water transportation and water filtration, and biomaterials development.

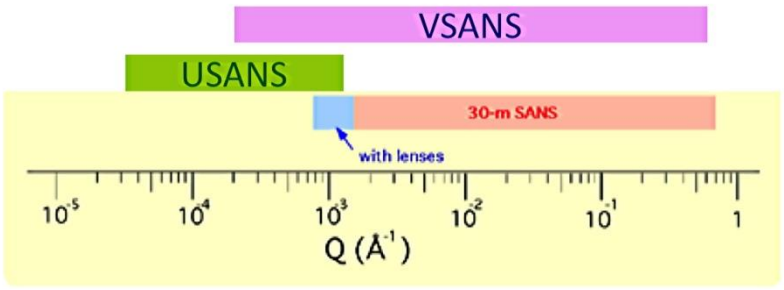
A fifth area of scientific opportunity is soft matter under confinement. Soft matter in porous materials, in thin films and membranes, and crowded in biological systems behave differently than in bulk. Examples of phenomena of interest include the structure of glassy materials and nucleation under confinement. Further understanding of these will impact energy production, microelectronics and communications, water treatment, and nanomaterials-based technologies.

The Essential Role of Neutrons:

Neutron techniques play critical roles in the study of these phenomena in polymer and colloid systems due to several characteristics of neutrons. Most importantly, large scattering contrast can be achieved by deuterating organic materials or using D_2O. For many structural studies, the changes in structure caused by the deuteration of a polymeric component is small. Ambiguity in structural determination can be reduced by the blending of labelled and unlabeled versions of the same component to match the scattering length densities of different parts of the sample, highlighting structural features of primary interest or providing for structural determination using the simultaneous analysis of multiple data sets. Neutrons are also typically far more sensitive to the locations and concentrations of low atomic number elements (e.g. H, D, Li) than are X-rays. In addition, neutrons are highly penetrating for many materials. This allows buried interfaces to be readily studied and also enables *in situ* measurements, not only under common conditions, but also under extreme conditions such as high pressure. This same penetrating nature can be further exploited for measurements of phenomena occurring inside a device during its operation (in operando). A common challenge with scattering techniques is a limitation on the range of scattering vectors available. With transmission mode "small-angle scattering" measurements with neutrons, construction of the VSANS instrument will provide ready access to scattering vectors values ranging over five orders of magnitude. For samples readily damaged by X-rays, the meV energies of neutrons allow nondestructive study, even for long measurement times. Finally, neutrons provide access to dynamics over multiple lengths and time scales not accessible with other probes. This capability ties in closely with structural studies, but is addressed primarily in the report of the Colloids and Polymers Dynamics working group.

Instrumentation needed

The most important single advance that can be made in instrumentation to open new vistas with polymers and colloids will be the installation of a new D_2 cold source providing a factor of two increase in flux. This is critical for enabling fast experiments with small beams. Future studies of structure in polymeric and colloid materials will exploit all of the following instruments/techniques: SANS, VSANS, USANS, Reflectometry (in particular, CANDoR),- Off-specular scattering (such as on MAGIK), neutron imaging and tomography, and neutron depth profiling (NDP). The recommendations summarized here focus on SANS-like and reflection geometry experiments. SANS and USANS cover close to five orders of magnitude in Q. SANS collection times are currently typically fractions of an hour while USANS



requires 5 to 6 hours to collect one data set with acceptable statistics. The VSANS instrument will cover the entire SANS range as well as the slow-to-acquire part of the USANS range. This will cut data acquisition for the enlarged Q range by a factor of two. Imaging and tomography will provide information on large scale structure, while NDP will complement morphological information with quantitation of chemical composition for important constituents present in small amounts (such as salts in corrosion).

A beam characteristic required for many advances is increased brightness. Increased brightness will enable measurements of kinetics with time scales of seconds or less (particularly for phenomena that are irreversible), the study of novel or unique materials in small samples (which can provide a more prominent role for neutron scattering in synthesis of new materials), and the study of temperature sensitive samples by leveraging the rapid temperature equilibration possible with small samples. Increased brightness also brings increased throughput with limited beam time (with combinatorial samples or in the context of the Materials Genome Initiative) and can enable resolution of position dependent structures in heterogenous samples. The use of multiple detectors also cuts measurement time. This will be done with the CANDoR and VSANS instruments, enabling measurements of kinetics. It is also possible to gain new capabilities beyond those possible just with brightness gains by using energy dispersive detection (e.g. CANDoR). For reflection geometry experiments the reductions in measurement time will be large. If some means could be devised for using energy dispersive detection for SANS experiments as well, this would further extend the range of time scales that could be studied.

Advances needed in sample environments, software, and preparation lab facilities

To seize the scientific opportunities described above, the following sample environment developments are needed for the SANS type instruments, listed in order of priority:

1. Grazing-incidence SANS (GISANS) capabilities on all SANS instruments.
2. Novel shear cell(s) operating under high hydrostatic pressure. Despite opportunities for significant advances, no neutron or x-ray facility has yet provided such capability. This will be a pioneering sample environment meeting critical needs of scientific communities.
3. High pressure cells with hydrostatic pressure up to 3kbar. This can be achieved by an upgrade of existing apparatus
4. Stopped-flow apparatus for time-dependent phenomena on the time scale of seconds or better.
5. Cells operating under time dependent external fields such as electric/magnet fields or pressure.
6. Shear cell(s) or rheometer for polymer melts with high viscosities.
7. Cells providing extensional or other more complex flow capabilities.
8. Apparatus for experiments with solid deformation such as tension or compression.

Even with substantial advances in sample environments, there will be muted impact unless enhancements are made in the software available to users. The software should be improved to provide real-time data reduction of large sets of data from rapid experiments, sophisticated (e.g. comparative) display of reduced data and real time preliminary data analysis. The greatest scientific and technological impact will be attained if these real-time capabilities can deal with the entire 2D data sets, rather than being constrained to consideration of slices or subsets of the data. Further development of offline data analysis tools is needed as well. A gallery of tools is needed, including analytical modeling capable of orientation determination from 2D patterns, inverse Fourier transformation, data interpretation based on computer simulations such as reverse Monte Carlo methods, and calculation of scattering patterns of objects or collections of

objects with arbitrary shapes that users can create by manipulation of those shapes in virtual space. New users in particular will benefit from ready access to a database of scattering patterns from previously studied structures.

Two additions of offline instrumentation for SANS are recommended. Provision of an in-house SAXS instrument would enable complementary X-ray scattering measurements that are very helpful in the modeling of data sets from complex structures. An offline rheometer would allow users to check the quality of colloidal and polymer nonequilibrium samples after transport to NCNR and before a rheo-SANS experiment.

We offer three recommendations for CANDoR, specifically. With its capability for rapid measurement, fully realizing productivity gains will require the implementation of some robotic instrumentation, for example for automatic sample changing for as many as 30 samples. Automatic sample changing will also necessitate rapid, robust auto alignment procedures. Since poor substrate quality is a common reason for the failure of user samples to properly exploit the abilities of the instrumentation, we recommend that prequalified substrates of high planarity and low microroughness be made available to users. The cost would be readily recovered in better utilization of the beam.

For reflection geometry experiments more generally we recommend the development of a sample environment for high pressure (up to 2 kbar) measurements at temperatures from 25 °C to 150 °C (for example for studies of supercritical fluids on interfaces). Here also there is a need for real time data reduction and display for rapid measurements, including the case of 2D data sets. Real time data fitting with common functional forms would be helpful for better utilization of the beam. Publication and dissemination of the results will also be strongly enhanced by the further development of software for offline data fitting. This software needs to be capable of handling a wide variety of scattering length density profile shapes including user-defined, parameterized functions, with a user interface that is friendly for the broader user community that does not have facility in scripting or coding.

Biological Systems (Structure)

Participants: John Marino (NIST, Chair), David Fushman UMD), Arne Gericke (Worcester Polytechnic), Richard Gillilan (CHESS), Frank Heinrich (CMU, NCNR, scribe), Ferenc Horkay (NIH), Mathias Loesche (Carnegie Mellon), David Worcester (UC-Irvine, NCNR).

Scientific Opportunities

High resolution structural biology methods, primarily x-ray crystallography and nuclear magnetic resonance (NMR), have yielded thousands of high-resolution 3-D biomolecular structures, which have provided unprecedented insights into biological function at the molecular level. Recently, structural biology has shifted its focus from structure and function of individual proteins to understanding how those proteins interact with each other and with DNA, RNA, lipids and other molecules in the cellular milieu. Among the highest impact structures currently being published in high-profile journals, like Nature, are membrane-bound proteins. But the general trend is that the highest-impact work now involves complex multicomponent systems that are very challenging to investigate with conventional techniques. In this context, neutron scattering methods have and can continue to make significant contributions by addressing problems where neutrons measurements provide unique insights into biological structure and dynamics in functionally and physiologically relevant environments. Some examples of research opportunities for neutrons include:

1) Conformationally dynamic and intrinsically disordered systems: a number of biomolecules (ribonucleic acids (RNA), oligosaccharides, intrinsically disordered proteins (IDPs), multi-domain, flexibly linked proteins) provide unique opportunities for neutron scattering methods, as these molecules do not yield readily to high resolution approaches and/or are potentially structural perturbed by the requirements of these techniques (e.g., inclusion in a crystal lattice).
2) Membranes and associated proteins: Neutrons provide the opportunity to study membrane systems and associated protein structures in native-like lipid environments. In these environments, proteins don't form the crystalline arrays necessary for X-ray diffraction studies.
3) Supramolecular complexes: Large multicomponent**,** molecular complexes (e.g. DNA/protein, RNA/protein and protein/protein) provide unique opportunities for studies using deuteration and contrast matching.
4) Biological imaging and In vivo structure: There is a potential for neutron applications in in vivo studies and imaging, although these are seen as longer term goals which will likely require new instrument development and biochemical tools (e.g. neutron specific contrast agents).

Neutron scattering methods are ideally suited for studies of these types of biological systems and could address important questions of structure in relation to: stability and aggregation (e.g. biologics formulation); solvation/hydration, ionic association with charged biopolymers (RNA/DNA), molecular crowding; conformational changes in response to environmental cues, post-translational modifications (e.g. glycosylation) and biomolecular folding and assembly.

Impact of the Recent Expansion of Neutron Capacity

The new instrumentation (VSANS and CANDOR) and updates to older instruments installed as part of the recent NCNR expansion provide significant new capabilities for addressing biological structure measurements. Some specific features and capabilities were identified:

VSANS

1. The expanded lower q range available on the VSANS (q = 0.0008 $Å^{-1}$ to 0.7 $Å^{-1}$) will be of great advantage as applications shift to larger biomolecular complexes and assemblies. This lower q limit exceeds commonly-available BioSAXS capabilities in the U.S. by nearly an order of magnitude, providing VSANS with a unique capability sought by the biological scattering community.
2. Higher intensity (x10) and smaller spot size enable smaller sample volumes and shorter measurements.

CANDOR

1. Higher throughput and shorter measurement enable systematic studies and reduce concerns about protein stability.
2. Higher sensitivity enables structure determination from lower surface coverage (at present, >5% needed for many applications) and enables the resolution of more subtle structural details (complexes, conformational changes).

Deuteration Facility (Phase I):

1. Provides biochemical infrastructure that facilitates access of the NCNR user community to perdeuterated and selectively deuterated proteins and RNA/DNA.

Future Needs and Opportunities

Instrumentation. As the NCNR biology user community grows, the capacity of the current SANS instruments, already oversubscribed, will not be sufficient to support the new users. New instruments are essential if this growth is to be accommodated. Biological samples, particularly deuterated ones, are expensive and demanding to produce. It is therefore desirable that these instruments be optimized for small samples, (e.g. 10 microliter volumes for SANS) and the highest achievable sensitivity. Improved neutron capabilities that allow time-resolved experiments were also seen an important for many biological application. In addition, an in-house SAXS instrument was seen necessary to provide an important complementary scattering technique that biological users would need. The NCNR could also consider support for mail-in service and remote data collection in the future, akin to synchrotron facilities nowadays. To this end, the subcommittee recommended:

1. A new Biological SANS instrument for NG5 with a higher effective neutron flux on the sample (e.g. Wolter optics) to accommodate smaller samples sizes and short-lived biological samples, and to reduce oversubscription and provide more rapid access. The instrument scientists hired to support the new proposed BioSANS instrument should have expertise/experience in Biological SANS applications.

2. A new SAXS instrument to enable users utilize their neutron beam time more efficiently and perform complementing SAXS and SANS measurements on identical samples. Also, it is necessary for users to have access to other equipment on-site that will assure sample quality before the experiment, analyze data during the experiment, and plan subsequent experiments in a timely and efficient way.
3. Improved SANS and reflectometry/diffraction instrumentation capabilities that allow more time resolved experiments.

Sample Environments: The subcommittee recommended the following sample environments be considered for development that would enhance the measurement capabilities particularly for biological samples:

1. In situ sample or on beam preparation and manipulation (e.g. dialysis, purification, size-exclusion chromatography (SEC))
2. Automatic sample change and manipulation (required for high throughput techniques)
3. Smaller volume sample cells (in most cases protein material is limited)
4. Lower background sample cells to improve signal-to-noise at high q for higher resolution.
5. Real-time complementary sample characterization at the beam or on-site (e.g. UV-VIS, surface plasmon resonance (SPR), electrochemical impedance spectroscopy (EIS), dynamic light scattering (DLS)).

Support Infrastructure and Facilities. Recent efforts have been made to improve the lab facilities at the NCNR. A Deuteration Facility has been established by NCNR in collaboration with the NIST Biomolecular Structure & Function group at IBBR, providing critical infrastructure to the biological neutron user community; however, additional efforts are suggested to further improve the infrastructure for biological measurements:

1. Deuteration Facility (Phase II): The addition of chemical synthesis capabilities in support of selective deuteration.
2. Expansion of the user lab capabilities to support biochemical sample manipulation and characterization. SAXS and light scattering are especially important for characterizing biological samples in solution in support of SANS experiments. Similarly, X-ray reflectivity and ellipsometry are essential for characterizing substrates before neutron reflectivity measurements are performed.
3. The hire of a full-time lab technician for managing biochemical support labs and, in the future, remote measurement / mail-in services
4. A laboratory in the guide hall for limited sample manipulation and evaluation that enables post-irradiation sample characterization by alternative techniques.

Data Analysis and Computational Modeling

Access and ease of use of data analysis and computational modeling tools continues to be an important area for improvement by the NCNR to support biological structure determination. The

NCNR has made significant strides in developing tools, like SASSIE, for modeling biomolecular structure restrained by neutron data using molecular dynamics and Monte Carlo simulations of molecular interactions and motions and constructing models for shape reconstruction. The NCNR could expand its support of the biological user community with respect to data analysis and modeling, including:

1. Cross validation of data analysis and modeling software developed at the facilities.
2. Development of tools for handling the increased need for large data set (2D) with respect to analysis and archivation.
3. Making the data analysis and modeling software more user-friendly for biologists.
4. Development of integrative biological modeling packages that incorporate other structure and scattering data with SANS/RD (reflectivity and diffraction from membranes).
5. Development of tools to link SANS/RD data with molecular computer simulations.

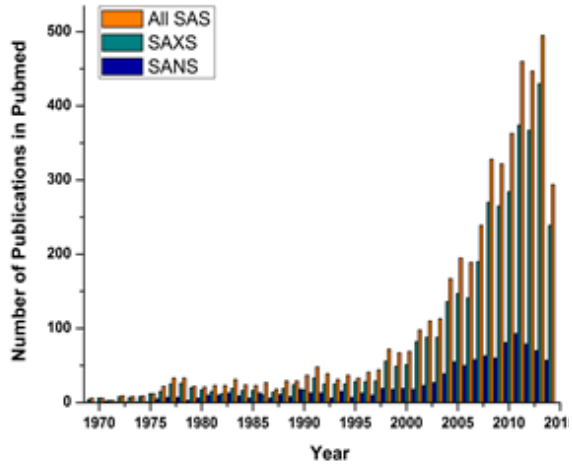
Expanding the User Base for Neutrons in the Biological Sciences

Despite a significant potential for biological neutron applications, the subgroup felt that the fast-moving field of structural biology is largely by-passing neutron scattering. Biological small-angle x-ray solution scattering has seen enormous and rapid growth in the past decade due to advances in algorithms and improvements primarily in sources and instrumentation. But most importantly, the growth of this field has been fueled by the ever-increasing importance of understanding the structure and function of multi-protein, DNA-, and RNA- complexes, which are generally difficult to crystallize. In contrast to SAXS, which has seen an exponential growth in application as assessed by peer reviewed publications, the usage of SANS leveled off in the past decade (see graph). The large base of experienced SAXS users that has developed in the field will increasingly need to take advantage of the unique properties of neutrons to resolve their structural questions. This is particularly true of molecular complexes containing components with differing average electron density (protein, DNA, RNA, lipids/detergents). In fact, it can be argued that neutron scattering (particularly contrast variation) is ideally positioned to advance structural biology in several key areas of increasing importance that cannot easily be addressed with conventional SAXS or other methods. To enable these advances, important barriers to education and facilities access need to be addressed. Expanding biological neutron scattering applications and growing the NCNR 'biological structure' user group remains a preeminent challenge.

Expanding the usage of neutrons in the biological sciences poses the following idiosyncratic challenges:

- Biologists have made enormous gains through refinement of traditional wet-lab techniques, but generally have little training in physical measurements and numerical data analysis at the level required for scattering experiments.

- Biological research moves fast; therefore, high-throughput and automated methods are favored.



SAXS and SANS publications listed in PubMed by year (as of Summer 2014) – courtesy of Kushol Gupta, UPENN

- Most biological scientists have a very different mind-set than physical scientists and engineers, and form a well-established community that is difficult to permeate

The panel expresses the opinion that the potential pay-off of neutron methods for structural biology are large, despite the listed obstacles, and suggests the following actions items to further increase the neutron user base in biology:

1. The NCNR should establish a Program in Biological Neutron Scattering with external partners. The panel recognizes that the NCNR in recent years added scientific personnel in key areas of bio-oriented research. Nonetheless, the addition of more staff experts is seen as essential. It was proposed that these experts be more formally organized into a NCNR coordinated research program. This program would drive neutron-based technology by developing neutron applications for biological applications. It would be essential for training a generation of neutron-literate biological scientists by integrating PhD students and post-docs into the day-to-day operations of the NCNR. This program might also help the broader biological community to identify experts in neutron methods and facilities for biology.
2. The NCNR much needs to continue to facilitate access for novice users and implement rapid-access procedures. To this end, a reorganization of the web site to include applications by research field rather than by instrument is proposed. In addition, it was suggested that feasibility time be more formally integrated into the proposal system.
3. The NCNR should continue and expand its outreach to the biological community. We suggest a series of focused workshops held in conjunction with major national meetings (e.g. Biophysical Society).
4. The NCNR should work towards better integration and partnership with x-ray scattering community. A suggestion was made to develop a mechanism of referral between user facilities, such as CHESS, where complementary SANS/SAXS would help address structural problems.

Dynamics of Soft Condensed Matter:

Participants: Yang Zhang, Sarah Woodson, Norman Wagner, Madhu Sudan Tyagi (scribe), Alexei Sokolov, Michihiro Nagao, Rob McQueeney, Ramanan Krishnamoorti (chair), Antonio Faraone, Bela Farago

Studying, understanding and modeling the dynamics of soft materials and their relationship to both the underlying structure and macroscopic properties remains a significant challenge. This challenge primarily arises from the need to access a broad range of energy (i.e., time) and momentum transfer (i.e., length) scales. Such research is central to a broad range of sciences and technologies, e.g. complex and multicomponent materials, development of the materials genome, as well as the development of hydrocarbon resources and novel energy generation and storage technologies. Measurement of soft materials dynamics is also proving valuable for researchers in human health and the quest to understand the function of the brain and the origins of brain diseases. Neutron spectroscopy provides an effective probe the time and length scales of the underlying motions important for function. However, the application of neutron spectroscopy to these problems is often limited by the size volume of the samples (several ml of sample) in current instrumentation. Reducing this minimum required sample volume is the first issue that needs to be addressed to broaden the use of neutron scattering methods to these emerging scientific and technological challenges in soft materials. There is also a significant over-demand for existing resources by the U.S. scientific community that must also be addressed.

Neutron spin echo, backscattering, and time-of-flight measurements of the dynamics are of significant value in soft matter science and are often unique in providing information on soft matter motion and kinetic processes on the nanoscale and on time scale from faster than picoseconds to microseconds. Consequently, there is a significant scientific benefit to be gained by integrating data and obtained from measurements of the dynamics of soft matter with data obtained from simulations, structural and other spectroscopic techniques. These data and metadata obtained from their analysis using standard and advanced modeling methods can be integrated into the materials genome initiative (MGI) database being developed at NIST for advancing the discovery, design and development of new soft materials.

Scientific Opportunity

Neutron spectroscopy offers unique and powerful methods to probe nano-to-micro scale dynamics on nano-to-micro length scales across a very diverse range of soft materials.

In biological systems, how intra-molecular motion correlates with function, and how local motion correlates with meso-scale properties such as aggregation, phase separation, diffusion and transport, remain outstanding issues. For example, the dynamical properties of intrinsically disordered proteins are essential for cell signaling, but may also contribute to protein aggregation, cell stress, and neuro-degeneration. These problems remain largely unaddressed. Other important problems emerging in biology include the dynamics of non-coding RNAs and chromatin in the cell nucleus, diffusion and transport of membrane proteins, protein aggregation in the brain and a broad class of problems associated with the elastic properties and lateral heterogeneities of lipid membranes induced by the incorporation of proteins, polymers and other molecules. In this direction, some advances have been made over the last decade thanks to quasi-elastic measurements using high resolution spectrometers such as backscattering and neutron spin echo.

These instruments have provided useful insights on how the softness in biomolecules essential for their function is achieved through motions between potential wells and within the single energy wells. Now, there is a significant scientific need to identify a "standardized" model protein to characterize and systematize a "universal" profile for protein dynamics through "round robin" characterization using a variety of spectroscopic techniques and provide a benchmark for theoretical and modeling of protein dynamics.

Understanding microscopic mechanisms of transport phenomena and collective dynamics of gas and liquid molecules, ions and protons through membranes, with and without external fields (e.g., electrical, flow and pressure), will have significant impact on water purification, gas separation, CO_2 capture, electrical energy storage and biofuel production. We need to unravel a connection between membrane dynamics and host matrix fluctuations with the transport of guest species, role of confinement and inter-molecular, Coulombic and other interactions. Collective dynamics is the key to macroscopic properties of glass-forming systems, ionic liquids and polymers. Estimates of cooperativity/heterogeneity length scale, their dependence on chemical structure and intermolecular interactions is crucial for understanding the dynamics and macroscopic properties of these materials. Special focus should be on bridging the local motions to continuum description by understanding dynamics at the mesoscale. This includes among others, Q-dependence of collective and incoherent dynamics, dynamics of nanoscale structures in ionic liquids and polyelectrolytes (single ion conductors), the nature of entanglements in branched polymers and in nanocomposite materials. Understanding the puzzling correlations between the fast dynamics (ps-ns time scale) and slow dynamics (e.g. biopreservation, fragility in glass-forming liquids) might provide better insights into the microscopic mechanisms of the slow dynamics, including aging.

Interfacial dynamics controls properties in nanocomposite and confined materials, liquid-liquid and liquid-air interfaces, and surfactant micelles. This includes water on various surfaces and liquid mixtures. Understanding dynamics in these intrinsically heterogeneous systems, the gradient of the interfacial effects, their dependence on interactions and degree of spatial confinement is critical for unraveling microscopic mechanisms controlling macroscopic properties of these materials. Moreover, many soft materials exist, are processed, or used in a nonequilibrium state, where the dynamics may be significantly different than at equilibrium. Examples range from polymer and surfactant solutions used in energy production to gels for cartilage replacement and membranes in fuel cells and batteries. Neutron measurements of molecular motion and transport in self-assembled systems under flow, applied stress, electric, thermal, magnetic, chemical or other fields will be of unique value in understanding the properties of such nonequilibrium materials.

Desired Neutron Instrumentation

a. There is a significant need to incorporate Wolter optics (or other methods) on all current and future neutron instruments to reduce beam size by focusing and therefore, dramatically decrease sample volumes necessary for the study of dynamics of biological and synthetic materials.

b. The investigations of dynamics in soft matter benefit from the use of long wavelength neutrons (5 Å and higher) resulting in improved resolution and access to longer time scale dynamics. The installation of the new deuterium cold source will significantly increase the

neutron flux for these wavelengths and benefit a number of instruments used for the investigation of soft matter dynamics. Hence this project should be given high priority.

c. The study of dynamics in soft matter can be significantly improved by the use of polarized neutrons, which can help separate coherent and incoherent contributions to the scattering most effectively. The Disk Chopper spectrometer can utilize He3 cells to provide polarization analysis capabilities, which would be useful to investigate coherent dynamics and collective dynamics in Q range that is typically inaccessible because of the dominance of incoherent signal.

d. The oversubscription of the NSE spectrometer has been identified as a significant impediment towards a better scientific understanding of the dynamics of soft condensed matter, especially on the nanosecond time scale. We therefore recommend the construction of a new state-of the-art neutron spin echo spectrometer. Two options are available: i) focus on the large length-long time scale dynamical processes with an instrument using long wavelength and optimized for using high field integral, with the goal of reaching microsecond resolution; ii) have high data detection rates with large detectors bank, the possibility of exploring high Q ranges to complement the capabilities of the existing NSE at low Qs. The soft matter community will probably benefit the most from the first option.

e. The current NSE spectrometer was recently moved to the new guide hall (less exposed to magnetic field perturbation due to the use of superconducting magnets on other instruments), equipped with a new polarizing cavity (polarization $\geq$ 97%), and new power supplies. Its resolution could be further improved with new precession coils and new correction coils. These improvements coupled with the upgrade in the cold source could push the instrumental resolution to the microsecond range.

f. There is also need to improve signal to noise ratio in backscattering spectrometer. This will help in separating slower relaxations (processes) from the instrumental background. This could be achieved by using a chopper directly in between the local shutter and the converging guide. This will need to be synchronized with phase space transformation chopper currently in use.

Sample Environment and Software:

The soft matter community, especially biological community, would significantly benefit from improvements to the available deuteration capabilities at NCNR.

To take full advantage of the advancement in data acquisition rates and resolution, comparable improvements in theoretical and modeling methods are necessary. State-of-the-art computer power is sufficient to match the length and time-scales accessible by the neutron scattering experiments; however such computing power along with theoretical expertise must be made available through NCNR to drive the development of understanding of the dynamics of soft matter. Further efforts in the development of new models and an iterative and interactive comparison with experiments are necessary. The user community will benefit strongly if these models are made available in user friendly software for *real time data analysis*. These models should also be helpful for users as predictive tool to identify challenges with their experimental design. Additional data tools such as multiple scattering corrections should also be made available to user community. These will lead to more efficient use of the neutron beam-time and shorten the time between experiments and publication. Appropriate software should be developed to allow for routine execution of experiments to study kinetic processes using SANS.

The community also requires further development of sample environments to study soft matter dynamics including non-equilibrium conditions:

- electric field for study polyelectrolyte membranes, batteries, fuel cells;
- magnetic field to investigate magnetic nanoparticle;
- shear and elongation flow fields with different geometry for understanding the microscopic origin of rheological behavior;
- pressure with different pressurizing mediums (e.g. CO_2, C_2H_2); higher pressures with the liquid cell to address free volume effects in glasses;
- UV-visible spectroscopy to introduce and independently monitor conformational change of proteins;
- air furnace and levitator for studying metallic glasses.

Materials for Sustainable Energy

Michael Toney, Andrew Allen, Rod Borup, Steven DeCaluwe, Greg Downing, Joseph Dura, Dan Hussey, Raul Lobo, Steven McIntosh, Michael Mendenhall, Vivek Prabhu, Howard Wang

Summary

Developing a sustainable, carbon-neutral energy infrastructure requires the development of new functional materials that are both inexpensive and high preforming and includes advances in solar generation, energy storage, catalysis, and carbon sequestration. This requires a better understanding of structure-function relationships, and, importantly, operation and degradation modes – watching materials and devices change as they operate or are synthesized. Such challenges exist in a range of energy technologies spanning generation (solar absorbers), storage (advanced batteries), and transformation (fuel cells) as well as carbon dioxide separation and sequestration, and subsurface fracture and fluid flow. Since the relevant materials properties encompass a wide range of length scales (from <nm to > mm), a multi-modal (technique) approach is required, and since studies during operation are essential, in-situ methodologies are necessary.

Neutrons have unique properties for probing matter that facility such in-situ, multi-modal material investigations, including high penetration, the ability to 'see' light elements, and being truly non-destructive. Thus, neutrons play a key role in the understanding and development of materials for sustainable energy. But to exploit and enhance neutrons for such research, new and expanded instrumentation and sample environments are indispensable. These include the ability to simultaneously measure a large Q range (from ≈0.001 nm^{-1} to >50 nm^{-1}), imaging with micron resolution, neutron reflectivity to few Å resolution, and dedicated (more specialized) powder diffractometers (high temporal resolution, high Q resolution, and large Q range). There is also a need (and opportunity) to develop complementary, multi-probe (multimodal) measurements, such as X-rays and neutrons. From the sample environment perspective, there is a need for enhanced, more diverse gas loading and flow capabilities (high temperature and pressure), better fluid flow environments, and enhanced sample handling. It is also important to develop an integrated methodology to use theory to guide the combined analysis and interpretation of neutron and X-ray data.

Introduction

Limiting the atmospheric carbon dioxide induced temperature rise is currently one of our greatest challenges. There are a number of approaches for developing a sustainable carbon-neutral energy infrastructure and these require the development of new functional materials that are both inexpensive and high performing. These span a range of usages for energy transformations (catalysts, solid-oxide and polymer-electrolyte membrane fuel cells), storage (Li-ion and other batteries), and generation (photovoltaics), manufacturing efficiency (concrete), for carbon dioxide separation and sequestration and more efficient subsurface fuel extraction.

Here we discuss how neutrons make an impact on this important research area and describe new instruments and environments that are needed to make a larger impact

on this research. It is imperative to note that these efforts require multi-modal and in-situ approaches. Hence, by the nature of this broad based research, no one single instrument will satisfy the research needs. A range of instruments and upgrades are needed, which we describe below, but which include the capability to simultaneously measure a large Q range, imaging with micron resolution, a neutron reflectometer with resolution of a few Å, more seamless overlap of data from different instruments, and more specialized powder diffractometers. We further explain the opportunity to develop a novel, complementary, multi-probe (multimodal) instrument. New and enhanced sample environments are needed for more diverse gas loading and flow capabilities, better fluid flow environments, and enhanced sample handling. The materials characterization community would also benefit from a theory guided, integrated methodology for the analysis and interpretation of combined neutron and X-ray data.

1.0 Important Scientific Opportunities

For the expertise of the scientists on this sub-panel, we focused on energy transformations (catalysts, solid-oxide and polymer-electrolyte membrane fuel cells, SOFC and PEMFC), storage (Li-ion batteries), manufacturing efficiency (concrete), carbon dioxide separation and sequestration and subsurface fossil fuel extraction. There was insufficient expertise to address issues related to combustion, nuclear materials and supercapacitors.

Neutron Measurements for Materials Design & Characterization
Hard Condensed Matter Physics

Summary

Neutron scattering, elastic and inelastic, continues to play a crucial role in advancing condensed matter physics as it provides a unique probe of the structural and magnetic correlations on a variety of length and time scales. This is reflected in the broad range of research areas of central importance to condensed matter physics and materials research that the Hard Condensed Matter Subgroup has identified, where neutron scattering is poised to make major contributions over the next decade. For example, transition metal oxides encompass many materials of both scientific and technological importance, including high-Tc superconductors, multiferroic and relaxor materials, thermoelectrics, and many more, where linked charge, spin, lattice, and orbital degrees of freedom must be elucidated. Systems with novel, exotic spin textures caused by geometrical frustration or by the formation of cooperative spin singlets provide unique laboratories for the exploration of the fundamental physics of many-body systems, but this exploration cannot proceed without full understanding of the static and dynamic spin correlations in these systems. Many of these unusual phenomena, and in particular the often strongly enhanced response to external fields that makes them of interest for potential applications, emerge in the vicinity of a quantum critical point, where long range order has been suppressed and where the ensuing competition between various ground states leads to low energy excitations that are uniquely suited for cold neutron scattering capabilities. It is also important to stress that hard condensed matter is a field that is driven by the continued discovery of new materials and novel phenomena with increasing complexity that involves coupled spin and lattice correlations. It is therefore reasonable to anticipate that new areas of applications of neutron scattering will continue to emerge, and thus it will be critically important to have state-of-the-art neutron scattering instrumentation capabilities in order to be able to quickly exploit new scientific opportunities as they emerge.

All of the research areas identified by the Hard Condensed Matter Subgroup offer unique challenges and opportunities that can be exploited by advances in cold and thermal neutron scattering instrumentation and accompanying advances in sample environments and polarization analysis. The greatest impact across many fields, in particular traditional condensed matter but also in topics of thin films, magnetic heterostructures and soft condensed matter, would be achieved with a new high wave-vector resolution, cold-neutron triple-axis spectrometer with a multi-energy, multi channel analyzer at the NG-5 end-guide position. As described in detail in the attached proposal, this instrument provides flexible energy and high momentum resolution, is compatible with polarized beam operation and high magnetic fields, and most importantly would provide *gains of more than two orders of magnitude in measurement efficiency*. These new capabilities, which would be not only unique in North America but in fact are not currently available anywhere worldwide, will enable future research at the frontier of condensed matter research as well as open cold neutron spectroscopy to new scientific fields, such as the exploration of small samples of newly discovered materials, the investigation of lattice

and spin dynamics in thin films and magnetic heterostructures, and investigations of strongly absorbing or "expensive" materials that cannot be produced in large sizes typical for neutron scattering.

Complementary to this new cold-neutron, triple-axis spectrometer, the development of an additional fully optimized modern thermal triple axis spectrometer with a multi-energy, multi-channel analyzer system and polarization capabilities compatible with high magnetic fields would provide further capabilities not currently available in North America, including options for resonant spin echo technique and magnetic form factor measurements. Continued improvements and advances in sample environment capabilities are also of great importance to ensure the ability to perform experiments at the forefront of condensed matter physics. Of particular interest are high magnetic fields and high pressures over a wide range of temperatures, and the possibility to utilize these sample environments with full polarization analysis. Because such sample environments are often restricted in positioning (particularly tilting), the development of a micro sample goniometer that allows alignment of samples or to perform out-of-plane scans is also highly desired.

In the following, we highlight a few of the scientific areas and opportunities that the Hard Condensed Matter Subcommittee identified as topics of central importance and we also include a detailed proposal for unique novel instrumentation that would guarantee the competitiveness of cold neutron spectroscopy at the NCNR with the potential for huge impact for the foreseeable future.

New Materials

Many of the topical problems in contemporary condensed matter science are derived from materials which either had not been crystallized 5 years ago, or whose properties were drastically underappreciated until recently. New materials are continuously being discovered and refined into a form which makes them amenable for advanced characterization. Neutron scattering has provided, and will continue to provide, definitive characterization information on these new materials, particularly when complex electronic, structural and magnetic degrees of freedom underlie their remarkable properties, as is often the case.

The importance of new materials with exotic physical properties to condensed matter science in general and neutron scattering in particular means that it is hard to predict exactly which materials and phenomena will be of most interest in even the medium term time scale. However it is clear and predictable that state-of-the-art neutron scattering infrastructure and sample environment, and a healthy and immediate interplay between materials preparation and crystal growth professionals with neutron scattering experts will be crucial to future successful materials research endeavors. An example is provided by the recent success and enormous impact that neutron scattering at the NCNR played in the discovery that high temperature superconductivity in iron based superconductors is strongly connected to magnetic fluctuations. The proposed new cold triple axis spectrometer with its more than two-orders-of-magnitude gain in measurement efficiency would be crucial to ensure readiness for similar success on new materials in the future.

As an example of a current scientific frontier, for which neutron scattering is currently severely limited but that the proposed instrument would enable to fully address, is the exploration of new electronic phases generated via the cooperative interplay between strong spin-orbit coupling and electron correlation effects. Examples of phases predicted to emerge within this new paradigm include new forms of high temperature superconductivity, quantum spin liquids, and topologically protected electronic states. Many of these new phases require a $J_{eff}=1/2$ ground state to realize, which currently is only possible via Ir^{4+} cations in a cubic crystal field. Neutrons would provide a pivotal role in exploring the physics driving the exotic properties of these materials. Iridium, however, is highly absorbing requiring that effective sample volumes are small. Additionally, the theoretical work surrounding these materials requires knowing *detailed* momentum information regarding both their static and dynamic spin behaviors in order to discriminate amongst a rapidly growing number of models. These two factors combined necessitate the need for high-resolution, high-flux neutron measurements in order to effectively explore the underlying ground states in these $J_{eff}=1/2$ systems.

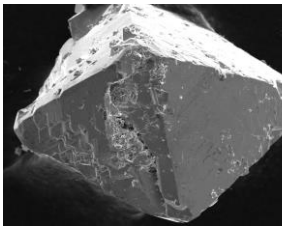


Fig. X: $Nd_2Ir_2O_7$ crystal, a candidate Weyl semimetal (~0.5 mm end

Linked degrees of freedom

Many novel properties of technological importance result from the competition and strong coupling of charge, spin, and orbital degrees of freedom. Examples include high-temperature superconductivity in copper-oxides, colossal magnetoresistance in manganites, large thermoelectric effects, e.g. in Na_xCoO_2, multiferroicity, ferroelectricity and relaxor behavior. The competition between different interactions often results in emergent complex states of matter not present in any of the Hamiltonians of the constituent interactions, but instead leads to emergent spin textures such as exotic chiral states, skymions, or self-organized inhomogeneous states. Furthermore, quantum fluctuations can sometimes prevent static ordering thus complicating the experimental characterization of the magnetic and electronic correlations.

State of the art neutron scattering simultaneously measures all the magnetic, lattice, and orbital correlations involved and therefore provides a uniquely powerful probe for such systems. The large variety of phenomena and associated varying states and energy scales, however, requires flexibility in energy and momentum resolution as well as in sample environment capabilities not currently available at the NCNR, or elsewhere in North America. For example, the complex energy landscapes usually encountered in these systems requires superb energy as well as momentum resolution in order to separate static correlations and to distinguish different excitations. Incommensurate order often encountered in these systems, particularly multiferroics, requires high wave vector resolution. Furthermore, many of the properties of interest in these materials are driven or strongly enhanced by external stimuli such as applied magnetic fields or pressure, making the availability of enhanced sample environment capabilities highly desirable for detailed studies. These investigations would also generally benefit from neutron polarization capabilities, e.g. cryopad / mupad devices, and with polarization analysis compatible with

significant applied magnetic fields (currently not possible) in order to distinguish the origin of measured correlations. For example the understanding of long-wavelength incommensurate magnetic orders may hold the key towards understanding exotic magnetoelectricity in multiferroics, a topic of great technological interest as it enables switching magnetic states with electric fields and *vice versa*. Of similar interest is the ultrahigh piezoelectric response observed in relaxor ferroelectrics, which is believed to be related to the presence of local, nanoscale regions of polarization. These "polar nanoregions" appear to condense from high temperatures and produce strong, temperature dependent diffuse scattering and neutron studies under high pressure would provide critical information on the nature of the polar nanoregions. But good energy and momentum resolution is essential to determine the origin of the diffuse scattering and its relation to unusual piezoelectric properties.

Unconventional superconductivity

Despite almost 30 years of intense research efforts, the mechanism of high temperature superconductivity in copper oxides remains poorly understood, though there is considerable evidence that conventional BCS theory is not applicable and that the pairing mechanism involves magnetic fluctuations. There are also many apparent similarities of the electronic phase diagram of the cuprates with other unconventional systems, such as heavy fermions and the recently discovered iron based superconductors, which further stipulate that superconductivity in these systems may all have the same origin. Neutron scattering directly measures the energy and momentum dependence of the imaginary part of the dynamical susceptibility, $\chi''(Q,\omega)$, and hence provides unique information crucial in addressing the relationship between magnetism and superconductivity. It is therefore not surprising that neutron scattering has made considerable contributions towards our understanding of unconventional superconductivity, including the observation that superconductivity in these systems is generally induced after long range antiferro-magnetic order has been suppressed by doping or pressure, the observation of coexistence of magnetic order and fluctuations with superconductivity, and the observation of resonant spin excitations below the superconducting transition as a signature of unconventional superconductivity in all these materials. The study of lattice and magnetic correlations and excitations in unconventional superconductors continues to provide unusual and surprising results, and would gain new perspectives with novel instrumentation, sample environment, and polarization capabilities. In particular, since superconductivity is a property arising from the generally poorly understood normal state, neutron scattering could reveal the ground state of these systems when superconductivity is eliminated by high magnetic fields or applied pressure. Advanced polarization analysis capabilities would further allow identifying the postulated presence of spin density wave or circulating current induced moments.

Quantum Critical Phenomena

One of the most important ideas to emerge in hard condensed matter physics in recent years is that of the quantum critical point (QCP). A QCP is a zero-temperature phase transition that marks a boundary in the space of ground states available to a physical system, and is traversed by tuning a parameter in the Hamiltonian such as spin-spin coupling in a magnet or carrier doping at a metal-insulator transition. Proximity to a

QCP usually strongly influences the physical properties at finite temperature, and the existence of QCPs is thought to underlie the behavior of many important materials, including the high-Tc superconductors and the heavy fermion systems. Various universal features and scaling behavior are thought to emerge near QCPs, and many of these should manifest themselves in the excitation spectra of the relevant systems at low energies. Consequently cold neutron scattering has a crucial role to play in exploring quantum critical (QC) phenomena, both in the systems mentioned above and in a broad class of insulating magnetic materials that provide clean model systems for exploration of QC phenomena. Further advances in the study of QC phenomena with neutrons will require increased access to both high magnetic field and high pressure sample environments (both at low temperatures) as these are the most convenient "knobs" the experimentalist can access to tune a system to its QCP. A further experimental challenge is that much of the universal scaling behavior near QCPs manifests itself not in conventional spin waves or other long-lived modes, but in the properties of strongly energy-broadened modes, often highly dispersive, thus requiring highly efficient instrumentation such as the proposed new multi-channel cold triple axis spectrometer.

Quantum spin liquids and other exotic magnetic spin states

Much current interest in the condensed matter science community is focused on materials comprised of interacting constituents whose pairwise interactions are inconsistent with the geometry of their underlying lattice. The best appreciated examples of this phenomena occur for materials which combine the tendency to antiferromagnetism and local lattice geometries made up of triangles and tetrahedra, but the phenomena are much more general than this as is demonstrated by the well known example of proton disorder in solid H_2O. Magnetic materials based on triangular, kagome, pyrochlore, and face centered cubic lattices often exhibit exotic low temperature states such as spin ice, spin liquid, and spin glass states which result from such competing interactions.

The main effect of frustration is to preclude the formation of a conventional ordered state at relatively high temperatures, so that the material has the potential to display an unconventional or exotic magnetic ground state. Such states are of intense theoretical interest. Their magnetic response is often characterized by unusual diffuse magnetic scattering and dynamics, reflecting the nature of the frustration which they display. In addition, lattice degrees of freedom can play an important role in the nature of the exotic ground states of these materials as lattice distortions can relieve frustration. A detailed understanding of magnetic and lattice scattering, as obtained from neutron scattering measurements, have the potential to allow detailed information to be extracted, thus allowing a unique determination of their exotic properties. Extreme sample environments such as high magnetic fields and applied pressures, alter the balance between competing interactions, either leading to "satisfied" states displaying long range order, or to new frustrated states.

Singlet Ground State Systems

Quantum fluctuations leading to a singlet ground state, which are not expected in a classical spin system, continue to attract interests of both theoretical and experimental physicists. Attention has been revived by the discovery of two dimensional S=1/2 spin

correlation in high Tc superconducting cuprates. Many new low dimensional magnetic systems with singlet ground states have been discovered, e.g. Haldane, spin Peierls, ladder, plaquette and dimer systems. The spin dynamics of these systems show a spin excitation gap from the non-magnetic singlet ground state to the first excited triplet state, which may be regarded as a direct consequence of the quantum spin fluctuations. More recently possible quantum phase transitions in these systems are attracting much attention. The non-magnetic ground state undergoes a phase transition to a classical magnetic state with finite sublattice magnetization by changing the relevant term of the system Hamiltonian, e.g. by applying magnetic field (~20 T) or pressure (~20 Kbar). Microscopically it is important to find out what the relevant exchange parameters are, and why the system behaves as a singlet ground state system. Neutron scattering is the method of the choice to obtain information on the microscopic scale.

Emergent topological spin textures

A field of research with considerable promise is the rich physics realized in helimagnets that host exotic phenomena with broader implications in fundamental physics such as skyrmion and magnetic soliton phases. Examples of systems with long wavelength spirals capable of hosting these unusual states include skyrmion states in MnSi and Cu_2OSeO_3 as well as soliton phases in $Cr_{1/3}NbS_2$ and related materials. Exploring the dynamics associated with the symmetry breaking inherent to these classes of materials provides a unique window into the electronic interactions driving their formation and provides a test for the proposed mapping of their properties within topological spin constructs (such as skyrmions). In particular, neutron scattering provides unique insight into the low energy spin dynamics as the length/time scales transition from the regime characteristic of the relatively large topological spin modes to those inherent of spins coupled at wavelengths shorter than those of the underlying spin spiral. High neutron flux at long wavelengths combined, critically, with the high momentum resolution are necessary to resolve dynamics away from the zone center and can potentially open a new field of exploration into these exotic magnetic states.

Thin films and magnetic heterostructures

Thin films and magnetic heterostructures are of interest for both technological applications and in the study of new physics. The development of polarized neutron reflectivity measurements has allowed the investigation of novel systems such as exchange springs, dilute magnetic semiconductors, and layered magnetic systems. However, thus far very little work has been done in inelastic measurements of magnetic thin films due to the extremely small sample volume. Such measurements, that would be enabled with the proposed new cold triple axis instrument, would permit determinations of the effects of strain on the exchange constants, for example, which are currently inaccessible through other probes. We should also note that as thin films are the product of nonequilibrium growth, some materials are only accessible in thin film form. For example, the GaAs:Mn dilute magnetic semiconductor cannot be cleanly produced in the bulk. However, in thin films it has been the subject of rich studies by neutron reflectivity measurements. Furthermore, it has recently been suggested by *ab initio* calculations that $EuTiO_3$ will be a multiferroic when strain is applied. It is however unlikely that chemical substitution will be able to produce the strains required, so such samples may only exist in film form. Another possible application of inelastic scattering to thin films would be in

the study of layered systems. For example, surface roughness could result in a change in the inelastic signal and it may also be possible to gain important insight into inter-layer couplings. Thin films also provide a new opportunity to study confinement and finite size effects. Finally, we note that the thin film community is one of the largest subgroups in magnetism. Working in concert with thin film growers to tailor materials for neutron study (for example, larger wafers) could substantially broaden the impact of neutron scattering.

Single molecule magnets and nanomagnets

Some of the recent exotic magnetic compounds under investigation are high spin systems (i.e. S = 10) and large molecules that contain magnetic atoms linked to organic groups. Many of these magnets have unusual low temperature properties stemming from the way in which the spins can interact. The scientific interest for the high spin entities is largely driven by the tunneling effects between excited states achieved in the application of magnetic fields. Under modest magnetic fields it is possible to shift transition levels relative to each other and observe tunneling effects and transitions between states. The use of spin echo techniques would be quite useful in determining the lifetime of the excitations. Other systems of interest include low spin organic systems. These are commercially quite important as they are soluble in organic solvents, can easily be processed and their properties can be fine-tuned by easily modifying their structure. They are also viewed as potentially important in quantum computing. One such example is Mn12 linked to acetate groups. The molecule contains an inner core of four manganese ions surrounded by an outer shell of eight manganese ions. Spin polarized experiments have been used to determine the overall spin of the complex in addition to locating exactly where the unpaired electrons responsible for the magnetism are located. Another example is V15 in organic groups where the V spins are frustrated due to the topology of the V lattice. Two possible ground states are present and the use of magnetic fields at very low temperatures will be very helpful in investigating the origin of the split ground state. At the same time, using single crystals it would be useful to resolve the nature of the spin states by using inelastic polarized beams.

INSTRUMENTATION

High Wave-vector Resolution Cold Triple Axis Neutron Spectrometer

As pointed out in the summary, a new high wave-vector resolution cold triple axis neutron spectrometer with multi-energy, multi channel analyzer installed at the NG-5 end-position would provide the highest impact across many disciplines and ensure future competitiveness of cold neutron spectroscopy at the NCNR. The detailed proposal attached to this report shows that this instrument achieves both high momentum and energy resolution that is highly flexible, and gains of more than two orders of magnitude in measurement efficiency compared with currently readily available instrumentation. We also point out that several components that could be utilized for this instrument, the monochromator drum and monochromator parts and crystals, are already available and in storage at the NCNR. This new instrument will also enable modern sample environment capabilities to be employed on the instrument, such at the 15 Tesla superconducting

magnet available at NCNR but which presently cannot be accommodated on any cold neutron instrument, and high efficiency—high intensity polarized beam capabilities.

Additional Triple Axis Spectrometer with full polarization analysis compatible with high magnetic fields

For a number of experiments, the triple-axis instruments still provide superior and unique capability, particularly compared to time-of-flight instruments, for inelastic neutron scattering measurements. While the BT-7 instrument at NCNR incorporates many of the new design innovations for triple axis instruments, its design does not allow polarization analysis with high applied magnetic fields, as this instrument is designed to utilize He-3 polarizer and analyzer close to the sample position, which is incompatible with magnetic fields. We further envision a new thermal triple axis instrument to incorporate the latest developments in multi-energy, multi-channel analyzers, which combined with the ability to perform polarization analysis would create a capability not available on time-of-flight spectrometers, while at the same time providing much greater measurement efficiency than conventional triple-axis spectrometers can provide. Performing spin-polarization analysis over a wide angle of scattered neutrons is not trivial, however, and should be incorporated into the spectrometer design from the start. Such a spectrometer would provide the capability to not only map out excitation spectra very quickly, as time-of-flight instruments can do, but would also be ideally suited for distinguishing the magnetic components of that excitation spectra. This can be extremely important in systems where magnetic and structural excitations are close to each other in energy/momentum space or where both spectra are changing as a function of some tuning parameter. Such a new instrument would also allow the incorporation of novel techniques, such as resonant spin echo techniques, as well as magnetic form factor measurements, which are currently not available in North America.

Proposal for a High Wave-vector Resolution Cold Triple Axis Neutron Spectrometer

Presently the NCNR operates two cold triple-axis spectrometers, MACS and SPINS. MACS is a new state-of-the-art spectrometer that views a dedicated cold source with a very large double-focusing monochromator system, to achieve the highest flux possible onto the sample [1]. To complement this measurement capability, the SPINS cold triple axis instrument can employ tight collimation to achieve high wave vector resolution when needed. SPINS was one of the first operating instruments included in the first guide hall project, and has been extraordinarily productive scientifically. However, presently it is the oldest neutron instrument in the NCNR portfolio (the BT-4 monochromator drum is older, but the FANS analyzer system is much newer). The instrument is still useful, but is antiquated and lacks capabilities that have been developed in the intervening years. In particular, it is now possible to greatly improve the neutron performance of a high wave-vector resolution instrument in terms of intensity, range of measurement, and flexibility. This can be achieved at modest cost as indicated below, and will also enable modern sample environment capabilities to be employed on the instrument, such at the 15 Tesla superconducting magnet which presently cannot be accommodated on any cold neutron instrument, and high efficiency—high intensity polarized beam capabilities.

Summary of Plan

We propose a new spectrometer, to be located at the NG-5 end-guide position recently vacated by the Spin-Echo instrument. The basic proposal is to use one of the new monochromator drums already purchased and stored on-site, together with the monochromator changer also constructed for the system. In addition, the PG for both the monochromator and analyzer is already purchased and available. We propose to construct a cantilevered sample table identical to the system on BT-7. For the analyzer, we propose to build a multi-channel-multi-energy analyzer system that will increase the capabilities by well more than an order of magnitude. A second option is to also construct a horizontally focusing analyzer system that is identical on the inside to the one on BT-7. Hence the amount of engineering services needed to implement this option would be minimal.

Detailed Proposal

- **Neutron Guide**
  - Replace the guide with a supermirror guide to improve the incident beam flux. Using an m=3 guide rather than the present Ni^{58} guide, together with designs such as ballistic transport, would increase the flux onto the sample by an order of magnitude.
  - The current NG-5 guide includes a neutron spin polarizer following the guide cut for the current SPINS spectrometer. We propose to put the polarizer on a two-position elevator so that we can have either unpolarized or polarized neutrons incident into the monochromator drum system. A cold Be filter option can also be installed in the guide path rather than after the monochromator as it is on SPINS, which will greatly improve instrumental background. We propose to obtain a new spin polarizer to

replace the current one, whose polarization efficiency could be greatly improved.

- **Monochromator Drum**
  - We propose to use one of the two new drums presently in storage. For the 'pipe' from the guide that leads through the drum to the monochromator, we propose to use the pipe currently on BT-7. This contains magnets that will be used for the polarized beam operation. The reason to do this switch is to construct a new pipe for BT-7 that has much thicker shielding laterally, to improve the radiation shielding at BT-7. The drum will be mounted on a gun-mount gear, identical to that on BT-7.

- **Monochromator System and Crystals**
  - We propose to use one of the double-stack monochromator systems already delivered and stored. We also have sufficient ZYA PG crystals in storage to mount on the monochromator system.

- **Sample System**
  - We propose to construct a non-magnetic cantilevered 'bucket' sample system identical to that on BT-7, with the exception of removing the underlying translation option and simply have a fixed position. The translation system will be replaced by a spacer to achieve the correct height for the goniometer. Given the 4″ higher floor to beam centerline, the serviceability of the gearing systems will thus be improved. Sufficient room will be available to readily accommodate all of the NCNR sample environment equipment, 4-circle option, and in-situ optically pumped He^3 polarization analyzer.

- **Analyzer System**
  - We propose to build a multi-energy analyzer, consisting of a series of PG crystals at a single scattering angle, with each blade set to detect a different energy. The number of blades is estimated at 40, but will require detailed engineering to set the number. Each blade will have a short 2° collimator to reduce the diffuse scattering from the crystal, and a single detector. This concept is a simple extension of the one already implemented on BT-7 using the 13 blade analyzer array and PSD (see Fig. 6f in [2]). We anticipate using a guide channel to eliminate the losses along the array, and increase the width of the blade with increasing distance from the sample/increasing final energy so that the full with of the channel is covered . A series of channels will be arranged horizontally to cover a wide angular range, similar to one of the proposed designs in the original BT-7 interchangeable analyzer document and implemented in the MACS double-crystal design. With the end-guide position, roughly half of the channels will be on the q-focused side of the instrument over the full energy range of the instrument (unlike MACS) in order to achieve the best wave-vector resolution.

- A second option is to build a second analyzer system identical in its operating characteristics to the one presently operating on BT-7. This is an extremely flexible system from an instrument capability viewpoint. The only proposed change would be to reduce the thickness of the poly shielding around the entire analyzer system, with the exception of directly along the sample-to-analyzer beam line so that no engineering changes to the collimator holders would be needed. We currently have sufficient ZYA PG in storage to mount on the proposed analyzer system. This will allow a maximum scattering angle of approximately 140° over the full energy range of operation.

- **Polarized Beam Option**
  - Neutrons can be polarized in the guide with the spin polarizer when that option is moved into position, and the polarized neutrons will be guided through the monochromator to the sample with build-in guide fields, identical to BT-7. He^3 cells will be used to analyze the polarization of the scattered neutrons for either analyzer option.

Possible Improvements

The above proposal outlines the new instrument at minimum cost. There are a number of improvements that could be implemented to improve the performance if engineering and funding is available, either in the initial stages of the project or at a later time, as follows:

1) The BT-7 analyzer system is designed so that other types of analyzers can be constructed and then easily interchanged. One option in the original BT-7 proposal was to have a diffraction-type wide-angle position-sensitive detector. This option became a low priority when the Materials Diffractometer was proposed for the new guide hall. Having such a PSD system as a third analyzer on this new spectrometer would recapture the measurement capability of the Materials Diffractometer.
2) Develop a spherical neutron polarimetry capability using µPad or cryopad. This would constitute a unique capability is the U.S.
3) In-situ optically pumping system for the He^3 cell used at the analyzer position. This would eliminate the time dependence for polarized beam measurements, greatly improving the accuracy and flexibility of such measurements.

Measurement Capability Improvements

The multi-energy-multi-channel system together with the m=3 guide will provide a measurement capability and sensitivity that is **more than two orders-of-magnitude** higher than the present SPINS spectrometer. The instrument will be able to provide an energy resolution 5X better than MACS, and a wave-vector resolution more than an order-of-magnitude better than MACS.

Presently the scattering angle (A4) on the SPINS spectrometer is restricted to ~120° at low incident energies, and is much reduced at higher incident energies (59° at 14 meV).

The new analyzer systems should allow scattering angles up to ≈140°, independent of the incident energy.
The 15 Tesla magnet, which can accommodate a 1.5 K stick or dilution refrigerator, can be accommodated on the new instrument, which is a sample environment system that would be most useful on a cold neutron instrument.
Analyzer shielding is greatly improved over that available on SPINS, which will improve the signal-to-noise for all measurements.
The state-of-the-art analyzer capabilities will greatly improve the measurement capabilities of the overall instrument compared to that of SPINS. For those types of measurements where the scattering of interest is localized in reciprocal space (i.e. traditional triple-axis measurement capability) the reciprocal space focus and flexibility of the second analyzer system will provide a dramatic improvement (as already amply demonstrated on BT-7) over SPINS. Nevertheless, the SPINS spectrometer is still quite useful, and can be continued in operation in order to provide NIST and outside scientists with an instrument to measure, for example, order parameters, check crystal alignment and align multiple arrays of crystals, and carry out other simpler elastic and inelastic measurements.

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The Future of Materials Chemistry at the NCNR

A Report from the "Materials Chemistry" breakout group at the "Neutron Measurements for Materials Design and Characterization" workshop

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Summary

Ours is an era of new materials. Innovative materials underlie technological breakthroughs in many areas of importance to societal wellbeing; the efficient harvesting of solar energy, new biocompatible materials in medicine, supersensitive detectors, lightweight alloys for jet engines and space exploration etc. Widespread and efficient adoption of new materials requires a new paradigm of progress by rational design. The White House's Materials Genome Project and the NRC's Frontiers in Crystalline Matter are two prominent examples that are now squarely in the focus of NSF DMR and DOE BES grand challenges of atomic control of materials. In the following sections we lay out a series of specific scientific and technological challenges where neutron diffraction and scattering can make major contributions. On the basis of these opportunities, we identify a critical need for the construction of a new high flux thermal neutron diffractometer (FAST), as well as upgrades to the existing high resolution diffraction capabilities (BT-1 to SHARP) and low energy inelastic scattering capabilities (DCS).

Assembling Functional Materials: Understanding Design and Synthesis

New neutron scattering capabilities can help enable the development of new materials by improving our ability to both *discover and rationally design* them. Crystal transformation, solid-state reactions, chimie douce methods of intercalation, and crystal assembly from liquids and/or clusters are all *dynamic processes* with mechanisms that

can be probed through rapid neutron scattering *in situ* in tandem with theory and high-throughput computation. One such process is the fast synthesis of Ti_3SiC_2 which proceeds through a previously-unseen intermediate discovered by *in situ* diffraction using D20 in 2002 with 1-second time resolution (Figure 1).

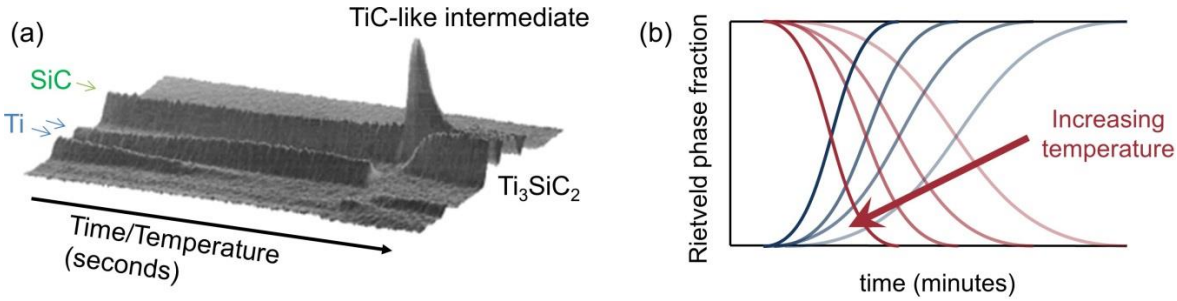

Figure 1. (a) 1-second data sets for a SiC + Ti reaction reveal an unpredicted TiC-like intermediate.[2] Mechanistic understanding of materials assembly demands probes with spatial, temporal, and chemical scales matched to actual synthesis conditions. In the schematic (b), Rietveld analysis of patterns acquired rapidly (requiring $Q \geq 8$ Å^{-1}) as a function of temperature allow an examination of kinetics.

There is tremendous opportunity for neutron scattering if an instrument at NCNR were built to allow the acquisition of Rietveld-quality diffraction data (allowing for refinement of oxygen vacancy concentrations and bond lengths, etc.) on the time scale of reactions (minutes), on sample volumes that are realistic for materials synthesis (~1 gram), at accessible temperatures (typically under 1000°C for metastable materials), and with versatile sample environments. The penetrating power, adjustable probe volume, and scattering contrast (especially *M*-O, *M*-H) of neutrons are a distinct advantage, while chemically-selective reactions can be probed in unique ways using isotopes.

Structural Complexity and Subtlety

Materials of scientific and technological interest, such as MOFs and other zeolitic solids, piezoelectrics, controlled thermal expansion materials, superconductors, ionic conductors, etc., frequently display a high level of structural complexity (very large unit cells) and/or subtle structural distortions. In many cases, the complexity and subtlety are integral to the material's interesting properties. In order understand the role of structure in determining properties, high resolution diffraction data is needed to resolve both subtle peak splittings and an adequate number of Bragg peaks at high *Q*, where overlap and loss of information is a major problem for complex materials. The current BT-1 instrument offers resolution comparable to that of any neutron diffractometer in the US, although far short of some high resolution instruments in other countries, but its count rate is too low. An upgrade to BT-1, which maintained or improved its resolution, while enhancing its count rate through better detector coverage would impact a wide range of materials studies by allowing for smaller samples and more extensive parametric experiments. Recent work on MOFs and many other systems at BT-1 serves to illustrate the great importance of materials with large unit cells and subtle structural distortions.

Development and Study of Complex Magnetic Materials

Magnetic materials have revolutionized human life, by enabling memory storage in computer hard drives, energy conversion and many other applications. A major strength of neutron scattering as a probe of magnetic materials is its unrivaled ability to examine magnetic ordering at the atomic scale. State-of-the-art neutron instrumentation is needed to understand the behavior and fundamental phenomena leading to complex or exotic magnetism, and help advance the development of new materials. The materials chemistry community in the U.S. is in need of a diffractometer with low background at low-*Q* (high *d*-spacing) for the study of magnetic materials that possess large magnetic unit cells, small magnetic moments, or both. We require an instrument with sufficient neutron flux and therefore a high signal-to-noise ratio at a *Q*-range down to approximately 0.2 $Å^{-1}$ (*d*-spacing up to 30 Å). New science on materials expressing exotic magnetism such as helical ordering, spin density waves, and skyrmions requires low *Q* data. Furthermore, many interesting classes of materials, including the parent phases of the iron-based superconductors and certain 5d oxides, can exhibit weak magnetism, with moments as small as a few tenths of a μ_B per magnetic ion (e.g. 0.25(7) μ_B in NdOFeAs[3]) which are difficult to resolve with current powder diffraction capabilities.

At the high end, we need the diffractometer to reach a moderate scattering angle of approximately 8 $Å^{-1}$ in *Q*. This metric is chosen for two reasons. First, due to the dropoff in magnetic form factors, there is relatively little information on magnetic structure at higher values of *Q*. Second, and perhaps more importantly, it is essential to be able to detect and quantify changes in the crystal structure that accompany changes in the magnetic structure, as described in more detail below.

Neutron powder diffraction is also a valuable technique for its ability to provide information on a material's crystal structure and magnetic structure *simultaneously*. This is important for systems where charge, orbital, and magnetic degrees of freedom are coupled, such as perovskite oxides and unconventional superconductors. To study phase transitions in magnetic materials where various degrees of freedom are changing with temperature, we require a diffractometer with high collection rates in order to analyze whole powder patterns at fine temperature steps. This will allow us to understand the coupling between spin and structural degrees of freedom; classify phase transitions as either first- or second-order in nature; and extract the critical exponent of the order parameter near the phase transition, which would allow us to classify the dimensionality of the interactions within the material. A successful example of such measurements is shown in Figure 2, where the magnetic and structural phase transitions in the oxides $NdBaFe_2O_5$ and $HoBaFe_2O_5$ were studied at the D20 beamline at the Institut Laue Langevin (ILL).[4] The rapid data collection is needed to see subtle changes in structure that arise from charge and orbital order that would have been missed by course temperature steps. Currently, these types of studies are impossible at U.S. neutron sources, and a new state-of-the art instrument at NIST would open up new areas for materials chemists interested in magnetism.

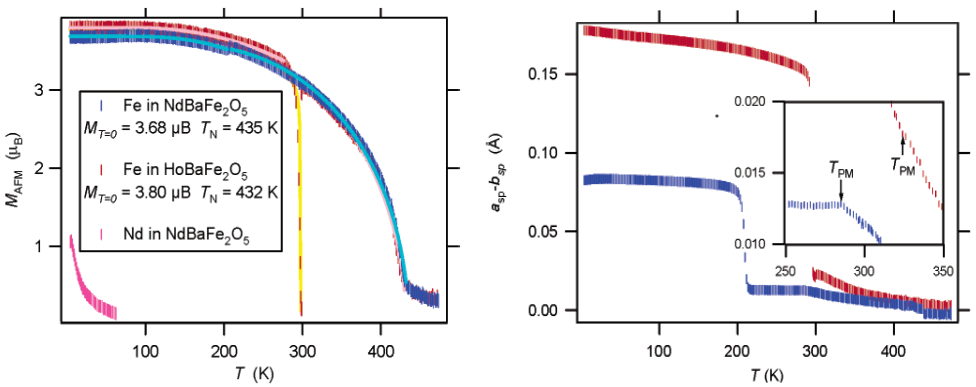

Figure 2. (a) The temperature evolution of magnetic reflections in $NdBaFe_2O_5$ and $HoBaFe_2O_5$. (b) Temperature evolution of a structural order parameter following the tetragonal-to-orthorhombic phase transitions.

In summary, to facilitate the development and study of magnetic materials, the US badly needs the capability to (i) collect data over the *Q*-range where magnetic Bragg scattering occurs, (ii) collect data rapidly to enable experiments where an external parameter (temperature, magnetic field, pressure) is varied over a fine grid, (iii) collect data that combines high intensity with low background, and perform polarized neutron experiments, so that weak reflections can be seen, and (iv) collect data over a sufficient range of *Q*-space and with moderate resolution so that structural features and magnetism can be followed simultaneously in the same experiment.

Structural and Magnetic Studies of Molecular Complexes

Magnetic anisotropy is the key component enabling applications ranging from permanent magnets to small-molecule data storage. Developing an improved understanding of magnetic anisotropy and furthering knowledge of structure-function correlations will enable the design of better molecules and materials. One method of parameterizing magnetic anisotropy is through axial zero-field splitting (*D*). Axial zero-field splitting in mononuclear transition metal complexes ranges from fractions of a wavenumber (~0.01 meV) to hundreds of wavenumbers (~10 meV), an extremely challenging energy range to access with conventional spectroscopy techniques. These transitions lie within the microwave regime, and tunable microwave sources are extremely rare, thus frustrating direct measurement of axial zero-field splitting. Prior research employing inelastic neutron scattering in polynuclear single-molecule magnets demonstrated zero-field splitting at the lower range of energies.[5] Applying this technique to the entire class of anisotropic mononuclear species would enable an improved understanding of magnetic anisotropy as it relates to electronic structure.[6] Neutrons offer an important advantage of a spectroscopic range not met by other techniques, and applying this to molecular solids will enable microwave range spectroscopic characterization of molecules. Similarly, vibrational modes in molecules can be

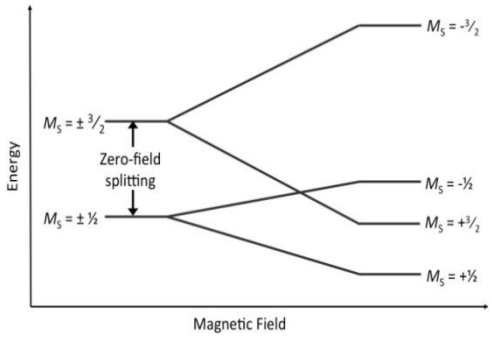

Figure 3. Diagram illustrating axial zero-field splitting, a key parameter of magnetic anisotropy.

investigated through inelastic neutron scattering. Molecular sciences are a key underutilized area for neutron studies that would broaden the impact of the facility.

Investigation of molecular species can be extended to atomic and magnetic structure measurements via neutron powder diffraction, particularly towards obtaining magnetic structures of molecules and differentiating between the positions of light atoms. This is potentially transformative since in most molecular systems magnetic structure information cannot be obtained. Neutron diffraction can also offer insight into dynamics of the light atoms by providing accurate atomic displacement parameters, not available with X-ray techniques.

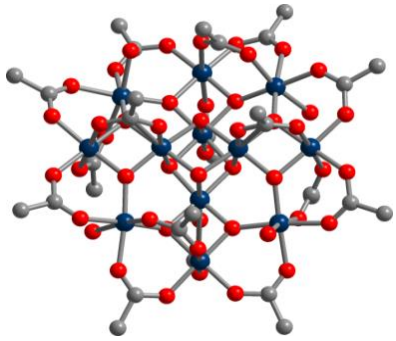

Figure 4. The single-molecule magnet, $Mn_{12}O_{12}(CH_3CO_2)_{16}(H_2O)_4$. Blue, red, and white spheres represent Mn, O and C atoms respectively. Hydrogen omitted for

Neutron studies of molecular systems have tremendous potential, yet current studies are frustrated by two key sample preparation limitations inherent to current neutron instrumentation. The first is the required sample size: a large scale synthesis of most new molecular species is on the order of 10-100 mg, far below the gram-scale size typically required for neutron experiments. The second challenge is deuteration to eliminate the incoherent background from ^1H. Deuteration of molecular samples is frequently cost-prohibitive since molecular compounds cannot simply be deuterated through soaking in D_2O, complex novel organic synthetic procedures incorporating new synthons must be developed. Devising new syntheses are exacerbated by the sample size requirements, as they necessitate not simply new synthesis but synthesis and scale up procedures.

Providing a high flux, medium resolution diffractometer would drastically reduce the sample size requirements. Demonstration of the viability of this approach has been performed at D20 at ILL where NPD data was acquired on mg scale samples.[7] Reduction of sample size requirements would enable the facile determination of magnetic structures and precision of light atom positions of molecules. Addition of powder polarization capabilities will, with sufficient statistics, permit extraction of spin-density maps, e.g. by maximum entropy data-analysis techniques. Similarly, significantly enhancing the flux on inelastic neutron instruments such as DCS would permit direct measurement of zero-field splitting parameters in molecular systems.

<u>In-Situ and In-Operando Studies of Functional Materials</u>

Moving from materials discovery to application is made more efficient by studies under conditions that mimic the end application. The time resolved study of pure materials, while changing temperature or chemical environment, can reveal important transient phases and kinetics. Better still, in-operando experiments, where a device or materials system are studied, can capture phenomena that may not occur for isolated materials. Neutron powder diffraction is powerful for both in-situ and in-operando studies, as it is a) compatible with a range of sample environments, b) sensitive to light elements such as O, Li and H that are frequently the key to function, and c) phase specific. It also samples larger (representative) volumes than x-rays, which can be beneficial in studies of larger industrial systems such as GE's NaNX batteries shown in Fig. 5. With current capabilities such an experiment requires tens of hours. This needs to be reduced to a few hours at most.

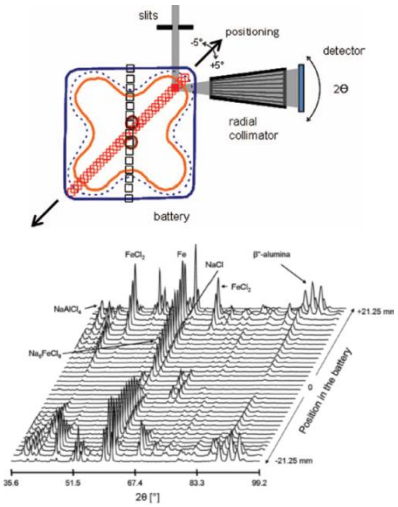

Figure 5. Spatially resolved diffraction provides data on the components in a GE NaMX battery.[1]

An instrument for efficient *in-situ* diffraction must have a high count rate while maintaining resolution ($< 10^{-2}$) and access to reciprocal space ($Q_{max} > 8Å^{-1}$) such that atomic level structure can be followed. It must accommodate a range of sample environments. Such an instrument would also be valuable for in-operando studies. However, as devices contain multiple materials, spatial resolution of a several mm^3 is desirable so that components can be interrogated independently. The development of a fast diffractometer at NCNR is clearly possible, as demonstrated by D20, but combining good spatial resolution with rapid acquisition will be a challenge. New in-situ and in-operando capabilities would enhance the development of materials for separations, batteries, fuel cells, catalytic membranes and many other applications. For industrial problems, such capabilities can help speed up materials selection and optimization by working directly at the system level under operating conditions.

Instrumental Needs

Three advances in neutron instrumentation are critical to addressing the above challenges: (1) a new, medium resolution, high flux, sample environment friendly powder diffractometer with user selectable polarization capabilities; (2) an upgrade providing higher count rates at the high resolution diffractometer BT-1; and (3) enhanced flux and polarization capabilities at DCS.

FAST

Significant new areas of materials research will be opened with the construction of a new high flux diffractometer (FAST). Required technical parameters are: access a Q range from (as low as reasonably achievable) to 8.0 $Å^{-1}$; Rietveld-refineable data collection time of one minute on a one gram sample with a simple analytically defined peak shape and a resolution of $\Delta Q/Q \sim 10^{-2}$; a low, ultrastable background; and copious sample space and appropriate materials of construction to support a wide range of sample environments (magnets, cryostats, gas flow with gas analyzers) and for the capability to

carry out polarized neutron experiments. This proposed FAST instrument is a natural evolution of the highly successful D20 instrument at ILL (for which the US has no comparable capability). No matter the specifics, the justification for a reactor source is clear: there are 1-3 orders of magnitude in signal to be gained simply by "leaving no neutron behind" (e.g. through improved optics and better detector coverage), and such a reactor instrument can readily provide simultaneous access to the low *Q* peaks associated with magnetic ordering and the higher *Q* peaks needed to precisely follow changes in nuclear structure, unlike the current generation of instruments at US spallation sources.

SHARP (BT-1 upgrade)

There is no substitute for high resolution neutron diffraction when elucidating subtle structural changes that underpin emergence in correlated systems or the performance of battery materials. As such, there is a continued clear justification for high resolution neutron diffraction vs. alternate (e.g. X-ray) probes for structure elucidation: the nuclear form factor does not decay as a function of *Q* allowing for precise refinement of atomic distances; the dependence of the contrast on nuclear configuration, rather than atomic number, is especially critical for observing light elements (Li, H, O) in the presence of heavy atoms (Ni, Ti, Bi, U); and direct sensitivity to magnetic order and magnetic correlations. The continued success of BT-1 demonstrates the long-term need of high resolution neutron diffraction at the NCNR. A modest investment in enhanced detector coverage could decrease the sample size and counting time requirements while maintaining or slightly improving the resolution that is already comparable to that of any other facility in the US.

DCS Upgrade

The Disk Chopper Spectrometer (DCS) has been successfully used in materials frontiers research, encompassing a vast array of scientific problems. In its current configuration, the Disk Chopper Spectrometer (DCS) is predominantly used for studies of low energy excitations including quasielastic scattering and diffusive excitations in a wide variety of materials. Because of its versatility, it has been heavily used by researchers across disciplines, from chemistry and biology, to soft and hard condensed matter to perform vibrational and magnetic spectroscopy and probe diffusion, magnetic excitations, etc. In the past two decades it has become a workhorse for inelastic scattering and has maintained a unique status for many years. However with the advent of spallation time of flight machines with similar capabilities, DCS has been seen to be less competitive because of its low flux even though it is only a factor of 6 down compared to CNCS (http://arxiv.org/pdf/1109.1482.pdf). With major upgrades to the neutron guide system, the increased flux from the cold source and repositioning of the detector bank DCS would again become extremely competitive on the national and international stage.

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Report on future opportunities for Engineering Materials at NIST

Don Brown, LANL, Sean Agnew, UVa, Craig Brice, NASA Langley, Timothy Foecke, NIST, Thomas Gnaupel-Herold, NCNR, Kavesary Raghavan, AK Steel, Aaron Stebner, Co. School of Mines.

The following summarizes discussion within the Engineering Materials sub-committee at the NIST Center for Neutron Research workshop, Aug 12-22, 2014. The discussion was aimed at next steps beyond the upcoming upgrades to the BT-8 diffractometer currently used for residual stress, texture and phase measurements.

With the development of high energy x-ray diffraction (HEXRD) techniques which can penetrate some materials to bulk depths, neutron scattering experiments relevant to engineering materials need to focus on areas where neutrons are currently and will continue to be unique. Specifically, Engineering Materials Neutron Scattering must rely on the ability of neutrons to penetrate high Z-materials while maintaining a large (>90°) diffraction angle. Also, engineering materials research with neutrons needs to capitalize on the sensitivity of neutrons to specific elements in contrast to x-rays, in particular, the ability of neutrons to sense hydrogen and other low Z-materials and the distinct contrast to unique elements as compared to x-rays (e.g. Co and Ni). Finally, engineering materials neutron scattering should capitalize on the ability to do bulk measurements without intensive surface preparation and the inherent infrastructure at neutron facilities to handle radioactive samples in order to carve out a niche in the area of nuclear materials. Going forward, identifying areas where opportunities exist, the committee recommends two distinct approaches; 1. traditional engineering diffraction measurements done bigger, faster, stronger and 2.) entering new areas of neutron scattering where engineers have not traditionally played.

Traditional Engineering Diffraction

For decades, engineering neutron diffraction measurements have used the advantages of neutrons to make measurements of residual stress, texture and phase in relevant materials, i.e. steels, aluminum, ceramics, etc. In particular, the ability to penetrate cm's into engineering materials while maintaining a high diffraction angle (>90°) allows the engineer to orient the bulk sample with respect to the diffraction vector (defined by the instrument geometry) such that the lattice parameter can be determined with sufficient spatial and d-space resolution in three orthogonal directions, typically two in-plane and one surface normal direction. These measurements are sufficient to calculate three orthogonal components of the stress tensor in a bulk component, which HEXRD cannot accomplish. Also, the typically large and relatively divergent neutron beam allows for more efficient grain sampling, resulting in quality statistical measurements of preferred crystallographic orientation (texture) and phase fractions.

Historically, the BT-8 diffractometer at the NCNR has been utilized to complete both residual stress measurements as well as texture and phase measurements to service its primary customers, but the optimization of these measurements are mutually exclusive. That is, residual stress measurements require very high resolution lattice parameter measurement, but are typically static, so throughput is not a dominant issue. In contrast, texture and phase measurement do not require high resolution, but do require time resolution to perform in-situ measurements where phase and texture evolve in real time.

The current upgrade to BT-8 will improve its ability to complete residual stress measurements. Specifically, the improved monochromator and detector will increase throughput, enhancing value to the customers. Moreover, the planned straining stages will add capability to study materials under more realistic conditions, e.g. multi-axial loading, again enhance value to the primary customers. To improve the performance of BT-1, the priority is to further increase the rate at which neutrons are counted at the detector, whether through increased incident flux (not clear how to accomplish), better optics, etc. The next most important upgrade would be to allow for manipulation of larger samples, such as large pipe sections. Finally, the ability to apply stimulus to these larger samples, such as stress or temperature, is desirable. The end goal is to study complex parts under relevant conditions of stress and temperature. All of these improvements are difficult to accomplish at BT-8 because of geometrical constraints.

Development of the BT-6 beamline could be used to complement the engineering materials program. Specifically, if the energy dispersive detector could be with neutron sensitivity from ~2Å - 4Å, with 1% or better resolution and mounted on BT-6, it could be used to service customers interested in phase fraction and texture in engineering materials (e.g. duplex steels). Specifically, one could imagine large parametric studies of phase and/or texture in steels as a function of processing or in-situ studies under processing or operating conditions. It should be noted that the engineering materials program at the SMARTS and HIPPO beamlines at Los Alamos has been very successful with this complementary model. The committee believes these directions will support the current customer base (NIST internal and industrial users) of the engineering diffraction program at the NCNR.

Novel Directions for Engineering Materials

With few exceptions, the study of engineering materials with neutrons has focused on diffraction measurements. Other neutron scattering techniques can certainly add value to engineering materials science, but it must be recognized that an outreach and education program would be necessary. Also, it must be recognized that engineering users work in a different mode than other sciences, e.g. physicist. The engineering user is interested in getting an answer to a specific problem and is less likely to understand scattering

techniques in depth and less likely to be a “career user” than their counterparts in a physics department.

The best opportunity for engineering materials science outside of diffraction seems to be awarded by taking advantage of the unique sensitive of neutrons to specific elements. For instance, neutrons have the unique ability to image hydrogen and this can be utilized to monitor hydrogen motion, for instance, to cracks in advanced steels, titanium, ceramics, etc, under stress. Also, neutrons differentiate between elements with similar Z-number in a way x-rays cannot. For instance, Co and Fe have very different neutron scattering and absorption cross sections enabling either imaging or SANS measurements of chemical separation or precipitation in high chrome stainless steels. Neutron reflectivity offers the ability to study near surface effects which are often quite important in engineering materials. These might include carburizing of surfaces which is used as crack prevention, or surface evolution on friction, or possibly near surface damage under ion irradiation. To enable such engineering materials research at NCNR innovative sample environments that can mimic the stress, temperature, quench rate, etc that materials experience during processing and use must be developed.

Significant interest was shown in the committee for simultaneous multi-physics and/or multi-length scale measurements on engineering materials. For instance, imaging hydrogen while doing diffraction to determine lattice strain/stress during deformation could be game changing. Because the geometry of the measurements might be different, one might have to consider multiple incident beam techniques to enable such multi-physics experiments. For instance, diffraction and imaging could be optimized independently by splitting the beam with distinct monochromators to bring the diffraction beam on sample at an angle of ~45° and the imaging beam on sample at 90°.

Finally, the committee recognized the unique niche to neutron scattering that radioactive or active materials offer. Neutron detectors are, in general, insensitive to background gamma radiation emitted by activated samples. Also, neutron facilities inherently have the infrastructure to handle even highly radioactive samples. Finally, because neutrons penetrate into the bulk, even in uranium samples, costly and hazardous sample preparation (such as polishing) is obviated.

Conclusion

To remain unique, engineering neutron measurements must be cognizant of the advances made in HEXRD and focus on the advantages that neutrons currently (and into the future) offer. These include bulk penetration, unique elemental contrast and sensitivity to low Z materials, and the ability to study activated materials efficiently. To strengthen the current engineering diffraction program efforts need to be made to increase the rate at which residual stress measurements can be completed on BT-8 while adding capability to

measure ever more complex samples in more complex environment, approach use and or processing conditions. BT-6 should be developed to provide complementary texture and phase measurements which are prohibitively slow on BT-8. To grow engineering materials in to new areas such as SANS and reflectivity, unique samples environments that allow in-situ heating, upset, quenching, etc must be developed in cooperation with the presumed customer.

Nanostructured Hard Materials

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Motivation and Scientific Opportunities

Magnetic nanostructures play a critical role in the storage of data. Even with the rise of solid-state flash storage in portable devices, in 2020 over 80% over the world's data (>3000 Exabytes) will actually be stored on magnetic hard disk drives locally or via the cloud. Modern magnetic recording devices are based on nanostructured magnetic particulate media and nanoscale multilayered magnetic sensors. Future technologies, including nanoscale bit-patterned media, thermal-assisted recording, and microwave-assisted recording, will push the limits of physical knowledge in magnetic nanostructures, interfaces, and spintronics. Additionally new solid-state memories based on magnetic random access memory (MRAM) cells are being actively developed to displace charge-based memories in critical applications. All these technologies are based on nanostructured materials where the accurate analysis of buried magnetic interfaces, domains, phase transformations and magnetic interactions is essential for progress.

Another important scientific opportunity is nanomedicine, and magnetic nanoparticles are expected to have great impact in magnetic resonance imaging, magnetic collection of disease marker proteins or circulating tumor cells for diagnosis, guided drug delivery, and magnetic hyperthermia cancer therapy. Currently, magnetic particles are known to cluster in solution, which is problematic for *in vivo* applications, though the acceptable cluster size varies with the application. Neutrons allow unique insights to behavior of these magnetic biomarkers and their interactions, both of which are critical for the advance of nanomedicine.

In addition to opportunities related to applications in information storage, sensing, and biomedicine, there are many more fundamental scientific opportunities that would benefit from using neutrons to study hard magnetic nanostructures. Neutron scattering is uniquely suited for quantitative depth profiling of magnetic multilayers. This measurement capability is further enhanced by the polarization capabilities at NCNR, which have already enabled high-impact magnetic studies. The unique decoupling of magnetic and structural scattering is a key strength of neutron scattering that differentiates it from other characterization techniques. These capabilities have already proven to be critical for understanding multilayers of complex oxides, where novel, spatially confined magnetic states can arise at heterointerfaces.

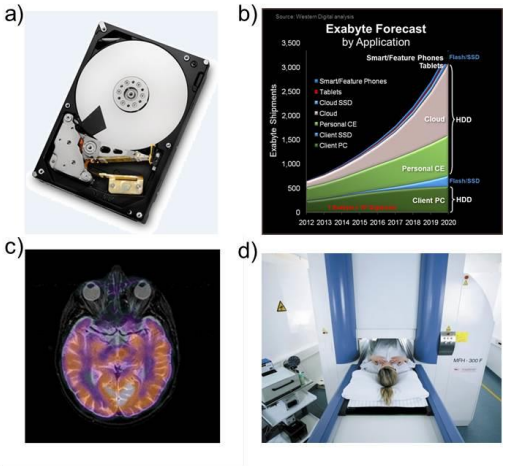


Fig. 1. Nanomagnetic materials play a critical societal role in data storage (a,b) and medical applications such as MRI imaging (c) and magnetic hyperthermia treatments currently being developed (d). (b) from Western Digital; (d) from Magforce Nanotechnology.

Though currently not feasible on nanoscale systems, inelastic neutron scattering enables measurement of low energy collective excitations, such as magnons and phonons. Similarly, magnetostatic interactions among lithographically patterned elements could lead to coupled low energy excitations. Neutron scattering is also a useful tool for investigation of magnetic configurations and spin polarization in exotic systems such as chiral carbon nanotubes, graphene, helical magnets, topological insulators, and flux lattices in new superconductors. Finally, neutrons provide the only probe of the multiple scales that are required for observing both small scale (within atomic distances of interfaces) and large scale magnetic structures. All of these fundamental investigations will draw new users to neutron scattering in the near term and will impact future technologies.

Research Opportunities Enabled by the NCNR Expansion

VSANS (Very Small Angle Neutron Scattering). The commissioning of the new VSANS instrument within the next few years on the NG3 guide is eagerly anticipated by the nanostructured hard materials community. Typical nanostructured samples (such as magnetic nanoparticles and nanowires) require access to large q ranges in order to probe correlations both within and among the structures. Currently the 30 m SANS instruments at NIST are capable of making measurements over scattering vectors ranging from q = 0.0008 Å^{-1} to 0.7 Å^{-1} (corresponding to length scales of approximately 1 – 800 nm). The UltraSANS (USANS) instrument covers the range from q = 0.00003 Å^{-1} to ≈ 0.01 Å^{-1}, but only with a 1-D detector. The new VSANS is designed to cover a q-range from 0.0001 Å^{-1} to approximately 0.7 Å^{-1} with 2-D detection, which substantially increases the length scales (up to ~6 μm) that can be characterized during a single measurement. In addition, the versatile optics of VSANS are anticipated to increase flux incident on the sample and to thus facilitate faster measurements of samples with limited volumes. This capability should open research opportunities for measurements of a *single* thin film or heterostructure with lateral inhomogeneities (e.g., from patterning or magnetic domain formation). Due to current intensity limitations on the existing SANS instruments, these measurements are only currently possible if a series of identical films can be fabricated and then stacked.

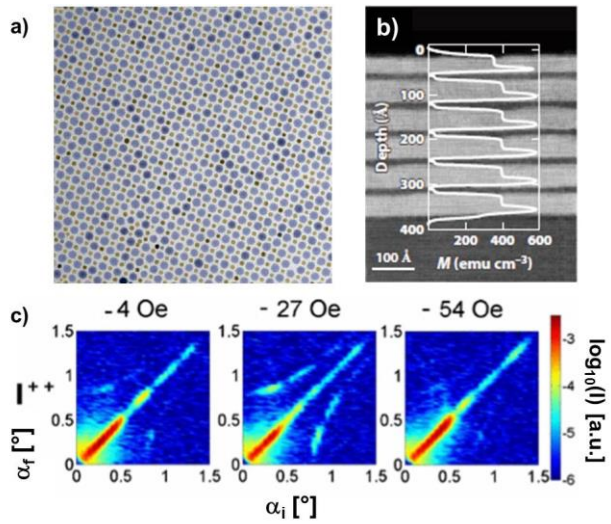


Fig. 2. The NCNR expansion will significantly enhance the capability to measure magnetic interactions between nanoparticles (a), resolve interfacial magnetism in oxide superlattices (b), and probe domain behavior through off-specular scattering (c). (a) from S. Majetich; (b) from S. May; (c) from K. Theis-Bröhl.

For many research problems of interest in the field of nanomagnetism, it is critical that VSANS has capabilities for polarization of both the incident and scattered beam. In addition, VSANS should routinely allow for versatile sample measurement geometries such as high-resolution grazing incidence SANS (GISANS) with optional polarization analysis. This

polarized GISANS capability will complement the *q*-range covered in off-specular measurements on MAGIK or CANDOR.

CANDOR. The new CANDOR instrument will enable several entirely new classes of experiments that have previously been impossible or impractical in the neutron reflectometry geometry. The combination of higher incident flux and multiplexing of both angle and energy analysis on the detector side will make CANDOR a uniquely powerful and flexible instrument.

The increased usable flux (at least 2 orders of magnitude) on the sample extends the practical *q*-range in reflectometry, when the high *q* limit is often imposed by the statistical noise when the reflectivity gets small. This is critical for increasing the depth resolution and enabling the detection of single-atomic-layer phenomena, including surface magnetism in topological insulators and other interfacial magnetism or interfacial doping effects. The increased flux also will allow rapid parametric experiments in which an experimental parameter is varied and the reflectivity is measured at each state. When a single fast scan of the reflectivity takes minutes instead of hours, users can measure depth-dependent hysteresis loops, moving phase boundaries vs. temperature or electric field, *in situ* annealing effects, and strain effects. It opens up a whole new field of reflectometry as a practical thin film material phase mapping tool. The multiplexed angular analysis (combined with a collimated incoming beam) will enable rapid off-specular studies of laterally inhomogeneous samples including materials with magnetic domains and phase-separated materials.

Recommendations for Future Instrumentation Development

There are many exciting opportunities on the horizon involving more intense, faster, higher resolution, extended *q*-range, multiplexed, and time-resolved neutron measurements with the possibility of more complex sample environments and pump-probe developments. The subcommittee recommends the following advances in instrumentation to open new scientific frontiers in hard nanostructured materials.

Wolter (focusing) optics would allow low divergence experiments, particularly SANS and GISAXS, to retain collimation without the need for pinhole optics and the huge loss of intensity that accompanies this. With nested, conical, Ni Wolter mirrors it is estimated that SANS could gain an average factor of 100 in flux – an increase that even the more powerful reactors could not be able to achieve due to thermal limitations. Such a huge increase in intensity would enable both the study of weakly magnetic systems and the increasingly-requested capability to study in-plane inhomogeneities on the order of nm to 100's nm (magnetic domains, quantum dots, magnetic correlation lengths, etc.). Characterizing the short-range, in-plane magnetic features of thin films is currently a capability that is highly desired by the scientific community, but can rarely be performed with current existing neutron flux.

Inelastic scattering enabled by a converging guide and multiplexing detectors holds the promise to transform the fundamental understanding of elemental excitations in reduced dimensions. Inelastic neutron scattering (INS) has played a crucial role in understanding excitations in bulk materials. Neutron inelastic signals are typically orders of magnitude weaker than elastic scattering, requiring more sample volume for an inelastic measurement. As such, INS measurements of nanostructured materials like nanoparticles and multilayers have historically been considered nearly impossible or highly impractical. Confinement

effects should have significant effects on the magnon and phonon dispersions of nanostructured materials, and characterization of these dynamics would profoundly contribute to fundamental understanding of condensed matter and potential device applications. Such dynamics have not been widely probed; if INS could be successfully applied to nanostructured materials, it would effectively open up an entirely new field of research: studying the effects of finite size, confinement, and interfaces for a multitude of materials. Inspired by recent developments, we recommend investigating the plausibility of developing an instrument for measurement of extremely weak inelastic signals. Focusing guide technology such as being employed on NG-C at the NCNR, combined with a multiplexing instrument similar to CANDOR could be a solution, although feasibility calculations are needed. One potential multiplexing geometry for enabling inelastic scattering would be a monochromatic incident beam with a detector array to simultaneously collect data over a variable energy range.

Cold polarized neutron diffraction would facilitate more direct measurements of spin configurations in thin films. Strain and interface effects can lead to magnetic structures (and phase diagrams) in films and heterostructures that are significantly different than their bulk counterparts. Magnetic neutron diffraction is a powerful tool for characterizing magnetic structure and the corresponding order parameters, field dependence, and coherence lengths. It is notable that high-angle diffraction is one of the only experimental techniques available to directly characterize antiferromagnetic order. Unfortunately, neutron diffraction from thin films presents special challenges. In general, peaks corresponding to magnetic order in the film are dwarfed by peaks corresponding to the underlying substrate and can be difficult to discern from background and intense nearby substrate peaks. Further, substrate peaks arising from subtle structural phase transitions or ordered defects can be easily mistaken for magnetic film peaks. This effect is particularly insidious, as such peaks can be temperature-dependent, increasing the possibility of mistaking them for magnetic peaks. We propose that the ideal instrument for studying weak magnetic diffraction from films would have maximized intensity, a position sensitive detector, utilize cold neutrons (to maximize angular separation between peaks at a given q) and full polarization capabilities to fully characterize magnetic structures. Such an instrument bears some similarity to the existing SPINS instrument, but SPINS is limited by a lack of q range, insufficient focusing optics to maximize intensity, and a lack of a position sensitive detector.

Time resolution on SANS allows measurement of morphology with sub-millisecond resolution. This is an extremely new capability with huge promise for the study of magnetic response to external perturbation (magnetic field, electric field, rheology, etc.), especially when combined with the vector magnetometry of neutron spin polarization analysis. Slower time scales (milliseconds) would promote the understanding of how magnetic nanostructures agglomerate and self-assemble in solution or under viscous flow within polymer systems. Faster time scales would allow meaningful interpretation of dynamic biological nanoparticle applications (hyperthermia cancer treatment, MRI contrast). Stroboscopic experiments could also be envisioned with real-time response.

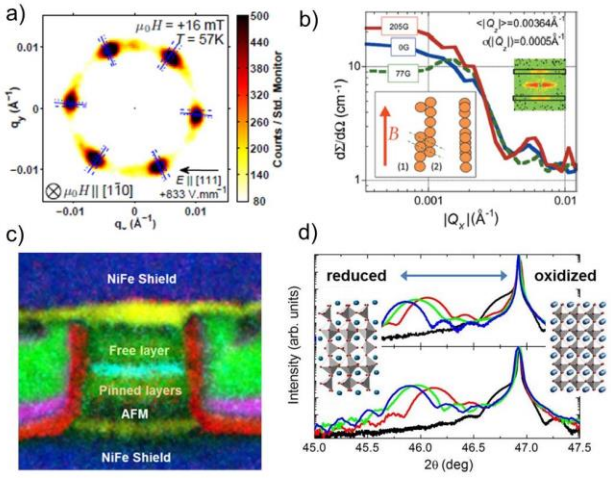
Vertical focusing mirrors on MAGIK. The vertical collimation required for GISANS measurement on the MAGIK reflectometer will restrict the incident intensity to a point that count times will be unreasonably long (weeks). A simplified Wolter optic would allow focusing of the beam along the normally-divergent direction perpendicular to the specular scattering plane. The beam is already tightly collimated in the scattering plane, so with the

envisioned focusing mirrors, a high-resolution GISANS measurement could be done simply by inserting the mirror into the beam path for part of the experiment. Full cylindrical Wolter optics have been demonstrated previously for a compact 2D SANS measurement. Therefore, this application of a simplified optic to focus only along a single direction (essentially taking a slice out of the center of a full Wolter mirror) has a high likelihood of success.

Direct inversion utilizes the nuclear-magnetic cross-term in conjunction with a built-in magnetic reference to recover the phase information lost in conventional scattering. This in turn, allows for a unique and unambiguous real space reconstruction for unknown structures. Such analysis has been very successfully applied to thin films and biological membranes using a magnetic reference layer within the reflectometry framework. We would like to see this extended to solution SANS where a magnetic nanoparticle reference attached at a single, known attachment point on the unknown object (for example a protein) would allow for direct inversion enabling real space construction of proteins, nanostructures, and other materials in solution. This advance also would resolve the issue of angular averaging inherent in solution science for multi-dimensional objects. The key will be to develop robust tags and single-point attachment appropriate for a range of sample sizes and shape.

Zero-field polarization analysis is not currently possible using the standard polarization set-up where a continuous magnetic guide field (0.5 mT or greater) is used to maintain the neutron polarization axis. However, even such modest applied fields can fundamentally alter the intrinsic magnetic morphology of interest. For example, solvated nanoparticles often chain and precipitate from solution when perturbed by an external field, while chiral spin structures and frustrated magnetic systems can also be modified in undesirable ways. To remedy this situation we propose the usage of Roger Pynn's newly developed CryoCup – a true zero field environment that can rival CryoPad at ILL, but at a fraction of the cost and space requirements. Its smaller footprint would enable multi-instrument shared use (reflectometry, SANS, triple axis) of zero-field polarimetry.

Recommendations for New Sample Environments



Sample environment plays a large role in the ability of users to extract crucial information on temperature, magnetic field, electric field, and stress driven effects as well as angular and time dependences. We have identified the following capabilities that would significantly enable the scientific opportunities at the NCNR.

Fig. 3. New sample environments would enable studies of electric field effects on skyrmion lattices (a), magnetic field effects on ferrofluids (b), industrially-relevant annealing conditions on magnetic multilayers (c), and changes in magnetization induced by *in situ* oxidation/reduction of oxide films (d). (a) from White *et al*., arXiv:1208.1146; (b) from Jain *et al*., J. Appl. Cryst. (2014); (c) image from HGST, a Western Digital company; (d) from S. May.

- High magnetic fields (12 T) compatible with polarized beam (possibly including compensating magnetic fields). This is useful for the investigation of superconductors, high anisotropy magnetic materials (ex. magnetic nanoparticles and exchange coupled materials), skyrmions, and to look for small magnetic signals such as surface magnetism.
- A new, more flexible sample staging area on the SANS instruments (higher q polarization analysis and higher field) to enable a high field magnet with sufficient space for polarization analysis.
- Simultaneous measurements of other physical properties:
    - electronic transport to correlate magneto-transport with magnetic structure and current-induced spin polarizations
    - optical/pump-probe in investigations of simultaneous metal-insulator and magnetic phase transitions and persistent photo-induced phenomena
    - oscillating magnetic fields (kHz-MHz, 10 - 50 mT) superposed on a dc bias field for magnetic hyperthermia investigations and investigations of magnetization domain dynamics
- Vector magnetometry for preparation of a particular magnetization state and for full magnetization analysis. This is useful for the measurement of magnetization components in directions other than the saturation direction and for graded anisotropy media.
- Wet cell and rheoSANS in a magnetic field: At the moment, the magnetic field capabilities are very limited and restricted to small fields. Construction of an electromagnet with sufficiently large openings is required.
- Application of large electric fields over large sample areas for studies of magnetoelectrics/multiferroics.
- *In situ* rotatable/translatable sample holders to enable sample motion for investigations of anisotropic effects and for accurate sample alignment.
- *In situ* strain application for investigation of the strain dependence of phase transitions.
- Rapid thermal annealing (RTA). RTA would enable *in-situ* methods to investigate phase transformation or sample modification so as to fully characterize industrially-relevant annealing processes on a single sample.
- Provision for gas flow to enable magnetic measurements of oxidized/reduced and intermediate states in complex oxide materials

- Complementary off-line equipment: SQUID, MOKE, PPMS to fully characterize samples, which can change between initial characterization at the user's institution and neutron measurement at NCNR.

Recommendations for Software Development

The new low-*q* measurement capabilities recently deployed and underdevelopment at the NCNR enable more efficient accumulation of data from nanoscale samples in part through the utilization of complex detection schemes. As a result, qualitative and quantitative interpretation of the resultant data is challenging and requires sophisticated data reduction and analysis software tools. The immediate software needs include:

- 2-D data fitting for SANS, in particular for polarized beam and rheology measurements
- 2D data fitting for GISANS and off-specular reflectometry
- Data reduction for CANDOR and other future multiplexing instruments