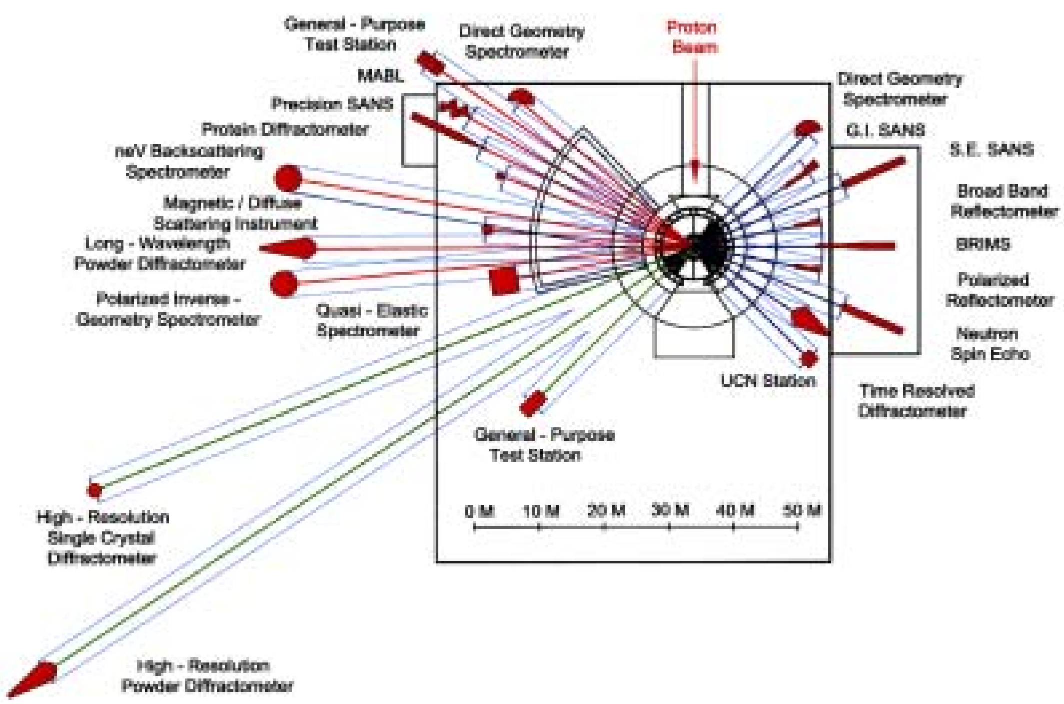
SNS 110040400-TR0004-R00

# Science Case for a Long-Wavelength Target Station for the Spallation Neutron Source




A U.S. Department of Energy Multilaboratory Project

SPALLATION NEUTRON SOURCE

Argonne National Laboratory • Brookhaven National Laboratory • Thomas Jefferson National Accelerator Facility • Lawrence Berkeley National Laboratory • Los Alamos National Laboratory • Oak Ridge National Laboratory

July 2002

## Acknowledgements

This is the final report to the National Science Foundation's Division of Materials Research on NSF-DMR 0073038, entitled "Conceptual Design of a Long Wavelength Target Station at the Spallation Neutron Source," awarded to the University of Tennessee. Dr. Thomas Mason, Director of the Spallation Neutron Source and Adjunct Professor of Physics, University of Tennessee, was the Principal Investigator.

The report was prepared by Dr. Lee Magid, Professor of Chemistry, University of Tennessee, and Acting Director of the Joint Institute for Neutron Sciences.

## TABLE OF CONTENTS

## Activities and Findings

**Research and Education Activities, Introduction**

This final report covers Activities and Findings supported by the grant in three areas: 1) development of the science case for a second, long-wavelength target station (LWTS) for the Spallation Neutron Source (SNS); (2) development of the technical concepts for the LWTS target, moderators, highest-priority instruments and data acquisition systems; (3) full documentation of the neutron user community's involvement to date in defining the scientific landscape that supports instrument performance parameters for the proposed LWTS and also for instruments funded and proposed for the High Power Target Station (HPTS), which will begin operation in 2006.

On February 2, 2001, one of the products of this grant, a phase I full proposal for the SNS-LWTS, was submitted to NSF-DMR from the University of Tennessee, with Dr. Thom Mason as the Principal Investigator. It quickly became apparent that this proposal represented a potential interagency partnership between DOE and NSF of such unprecedented size and complexity that it raised important policy questions as well as practical issues related to implementation. Therefore, the proposal was administratively withdrawn.

In Spring, 2001, the Interagency Working Group (IWG) on Neutron Science of the Office of Science and Technology Policy began its work. The IWG conducted an in-depth review of the status of exisiting and under-construction U.S. neutron scattering facilities, including their construction and operations schedules, facility budgets, staffing levels, neutron measurement capabilities, and the scope of their in-house science programs and external user programs. The IWG recommended, in a report issued in June, 2002, that:

"The highest priority for federal investments in neutron scattering is to fully exploit the best U.S. neutron source capabilities – including the SNS – for the benefit of the broadest possible scientific community.

The steward agency for each of the major neutron facilities form partnerships with other federal agencies for the purposes of meeting the objectives of the first recommendation."

The IWG's first priority for applying these recommendations states:

"The Department of Energy, the National Science Foundation, and other interested agencies should immediately establish a framework for an interagency partnership to provide funding resources to develop and operate a robust suite of instruments, approximately 75% of full instrumentation, to address a broad spectrum of neutron scattering measurements at the SNS. To be timely, the framework for instrument development should be effected within the next six months."

It is reasonable to assume that DOE will view eventual construction of a second SNS target station as their responsibility, given their role as steward agency for the SNS. Thus the priority quoted above concerning interagency cooperation on a robust instrument suite may cover in the long term both the HPTS and the proposed LWTS. Given that context, the results from this grant - the science case development, the LWTS technical concepts report and the documentation of scientific input into the instrument suites for both the SNS-HPTS and the proposed LWTS - provide a collective roadmap that the scientific community can use as a resource in preparing funding proposals for instruments on either target station.

**Science Case Participants**

The grant supported a core group of scientists from academic institutions, coordinated by Profs. Lee Magid (Tennessee) and Henry Glyde (Delaware). With the help of several scientists at Argonne National Laboratory (ANL), they developed the science case for the LWTS. They engaged the broad user community for long-wavelength neutrons - both current and potential users - through a series of focused workshops during the period 2000-02 and at breakout sessions at the general SNS users' workshop in

May, 2000. This community includes neutron scatterers as well as scientists interested in fundamental physics using neutrons. This group also engaged neutron scientists beyond the core group to help with writing various sections of the Science Case that appeared in the full proposal, phase I, for the SNS-LWTS.

The science case participants had frequent meetings, both separately and jointly with the group working on the technical concepts. The focused workshops convened by the science case participants included:

| Working Group | Organizers | Location/Date |
|---|---|---|
| Soft Matter | J.K. Blasie, R. Briber | Univ. Maryland, Apr. 19, 2000 |
| Magnetic Materials | C. Broholm, D. Argyriou | ANL, Apr. 27-28, 2000 |
| Disordered Materials | H. Glyde, C-K Loong | Univ. Delaware, Apr. 28-29, 2000 |
| Crystallography | A. Wilkinson, J. Jorgensen | ANL, May 12, 2000 |
| Chem. Spectros. and Dynamics | H. Bordallo, J.K. Blasie | ANL, Oct. 10-12, 2000 |
| Structural biology | C. Dealwis | Univ. Tennessee, Dec. 18-19, 2000 |
| Vibrational Spectros. | J. Larese | Knoxville, TN, Feb. 1-2, 2002 |

**Appendix A** provides reports for most of the workshops. Attendance at these workshops varied from 13 to over 40 participants. In each case, instrument scientists from the SNS project were in attendance to assess the needs of the user community and to provide feedback on ongoing instrument design. The first three workshops occurred before the grant began on May 1, 2000; Per agreement with Dr. Tessema, support for the participants' travel and subsistence was provided by NSF-DMR 9819471 (L.J. Magid, PI). The current grant provided support for the three remaining workshops. Reports from the workshops are available upon request.

Each of the working groups also held a break-out session at the SNS Users' Meeting, held May 22-24, 2000 in Washington, DC. Partial support for the attendees was provided by the grant.

A joint meeting was held at ANL from January 25-27, 2000. At that meeting, the science case participants and the instrument scientists prepared a short report outlining their agenda for interaction, as well as a first strawman set of instruments for the LWTS. This list, with modifications, was translated into conceptual designs for high-priority LWTS instruments. These concepts can be found in the report entitled "Technical Concepts for a Long-Wavelength Target Station" [report numbers ANL-02/16 and ORNL/SNS-TM-2001/163], which has been uploaded as part of this Final Report.

On Sept. 15, 2000 a subset of the science case participants and of the target/moderator and instrument teams met at ANL to review the schedule for developing the phase I (science case) proposal to NSF and for completing the Technical Concepts Report. The schedule for submission and review of the phase I proposal was negotiated with Drs. Weber and Haworth of DMR by Thom Mason (SNS; UT) and Lee Magid (UT) on Sept. 13, 2000.

At the Sept. 15 meeting, an outline of the phase I proposal was developed, and section leaders were asked to prepare first drafts for review at a meeting on Oct. 23, 2000 at ANL. Section leaders included: D. Argyriou (ANL), J.K. Blasie (Penn), H. Bordallo (ANL), R. Briber (Maryland), C. Broholm (Johns Hopkins), J.M. Carpenter (ANL), C. Dealwis (Tennessee), H. Glyde (Delaware), J. Jorgensen (ANL), C.-K. Loong (ANL), L. Magid (Tennessee), T. Mason (SNS-ORNL), H. Myron (ANL), J.W. Richardson (ANL), M. Snow (Indiana), A. Wilkinson (Georgia Tech).

During November and December, 2000, the phase I proposal went through several iterations, with input being sought from all members of the various working groups. On Nov. 20-21, 2000, there was a meeting held at ANL to review both the science case and instrument concepts in the draft proposal. Approximately 100 people attended. In addition to presentations by leaders of the various groups preparing the proposal, the attendees heard from Dr. David Moncton, Executive Director of the SNS project, Dr. Patricia Dehmer, head of DOE-BES and Dr. Thomas Weber, director of NSF-DMR.

As described in the Introduction, the phase I proposal was submitted to NSF-DMR on February 2, 2001. It was later administratively withdrawn.

*Plan for Educational Activities.* The phase I proposal also detailed a K-20 educational outreach program proposed for LWTS, called the Neutron Sciences Training and Education Center. It was developed by Dr. Harold Myron, ANL's director of educational programs, in collaboration with Dr. Linda Cain, formerly his counterpart at Oak Ridge National Laboratory. Dr. Cain's role is now being filled by Dr. Al Ekkebus, coordinator of user programs for the SNS project.

*Subawards to Science Case Participants.* Four subawards were made with the permission of NSF-DMR, and several additional science case participants were paid as consultants for their work. Final reports are found in **Appendix B.** Dr. D. Mikkelson of the Univ. of Wisconsin-Stout developed a prototype remote data access and visualization system for LWTS instruments. Dr. A. Wikinson of Georgia Institute of Technology was funded for development of the scientific case for powder diffraction at LWTS, for workshop organization and for participation in related discipline-based workshops where he presented information on SNS instrument concepts. Dr. H. Glyde of the Univ. of Delaware was funded to serve as a science case coordinator, to organize workshops on the scientific case and neutron-scattering instrumentation for disordered materials and glasses. Dr. B. Heuser of the University of Illinois at Urbana-Champaign was funded to perform neutronics calculations to determine the utility of a pelletized moderator for the LWTS. This work was performed to support the technical concepts team looking at LWTS target/moderator systems.

## LWTS Technical Concepts Participants

The grant also supported a group of scientists at ANL, via a subaward from the University of Tennessee, who developed the concept for the LWTS target and moderators, led the effort on design and prioritization of LWTS instruments, and compiled the respective performance cases. Jack Carpenter, technical director of Argonne's Intense Pulsed Neutron Source (IPNS) and PI on the subaward, led the target/moderator group; Jim Richardson, group leader, IPNS Neutron Scattering, led the instrument scientists.

The target/moderator group met frequently throughout 2000 and early 2001. Periodic updates from these meetings were distributed electronically to all participants in the LWTS collaboration; these updates were provided in the 2001 Annual Progress Report on this grant. At the joint meeting with the science case participants in Jan. 2000, a professional review of the pre-conceptual target station design occurred, conducted by Drs. Bauer (PSI-SINQ), Broome (ISIS), Russell (Los Alamos) and Watanabe (JAERI). Their report is available upon request.

The target/moderator group and the instrument group wrote the sections in the full proposal submitted to NSF-DMR on Feb. 2, 2001, dealing with the LWTS Technical Concept and with Proposed Instrumentation. This material, in updated format, also appears in the report entitled "Technical Concepts for a Long-Wavelength Target Station". [It appears in a separate .pdf file, uploaded as part of this Final Report.] As noted in the report, the instrument design concepts are the joint work of the science case participants and the technical concepts participants. Of the 20 LWTS proposed instruments in Fig. 2.2 of the report, eleven concepts are discussed in some detail. Taken together with the instruments approved and funded for the SNS-HPTS, approved and seeking funding, or at the concept stage, a robust suite of instruments to address the needs of the scientific community at SNS is on the horizon.

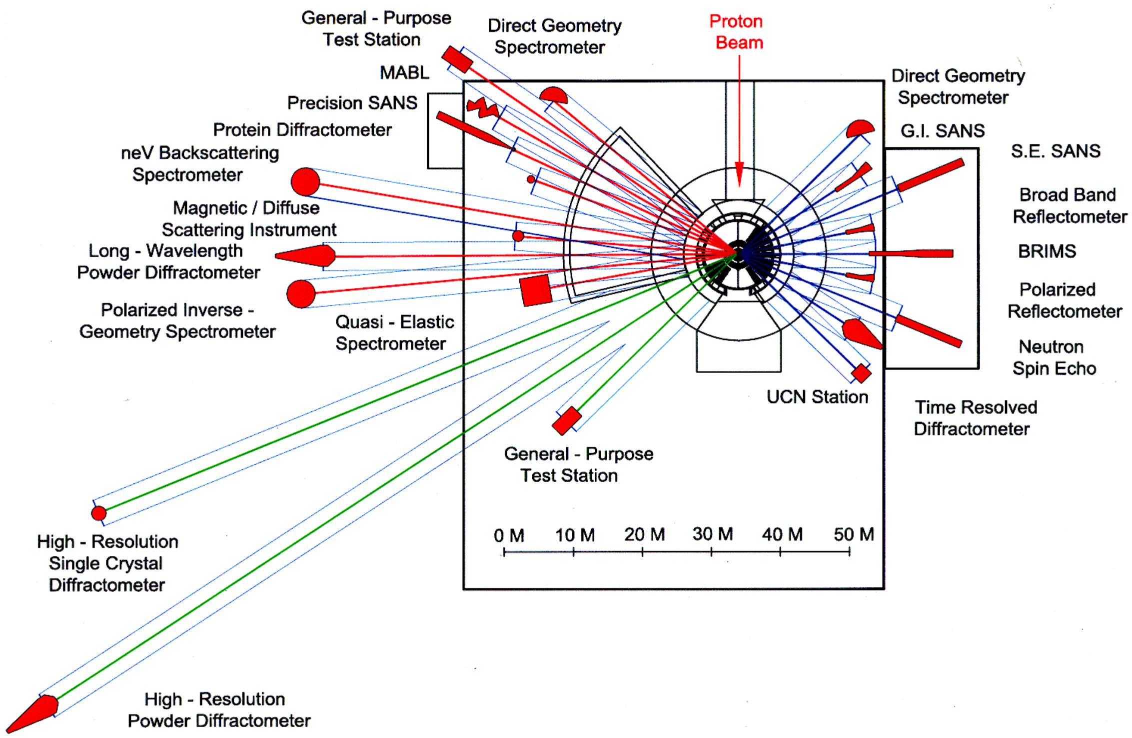
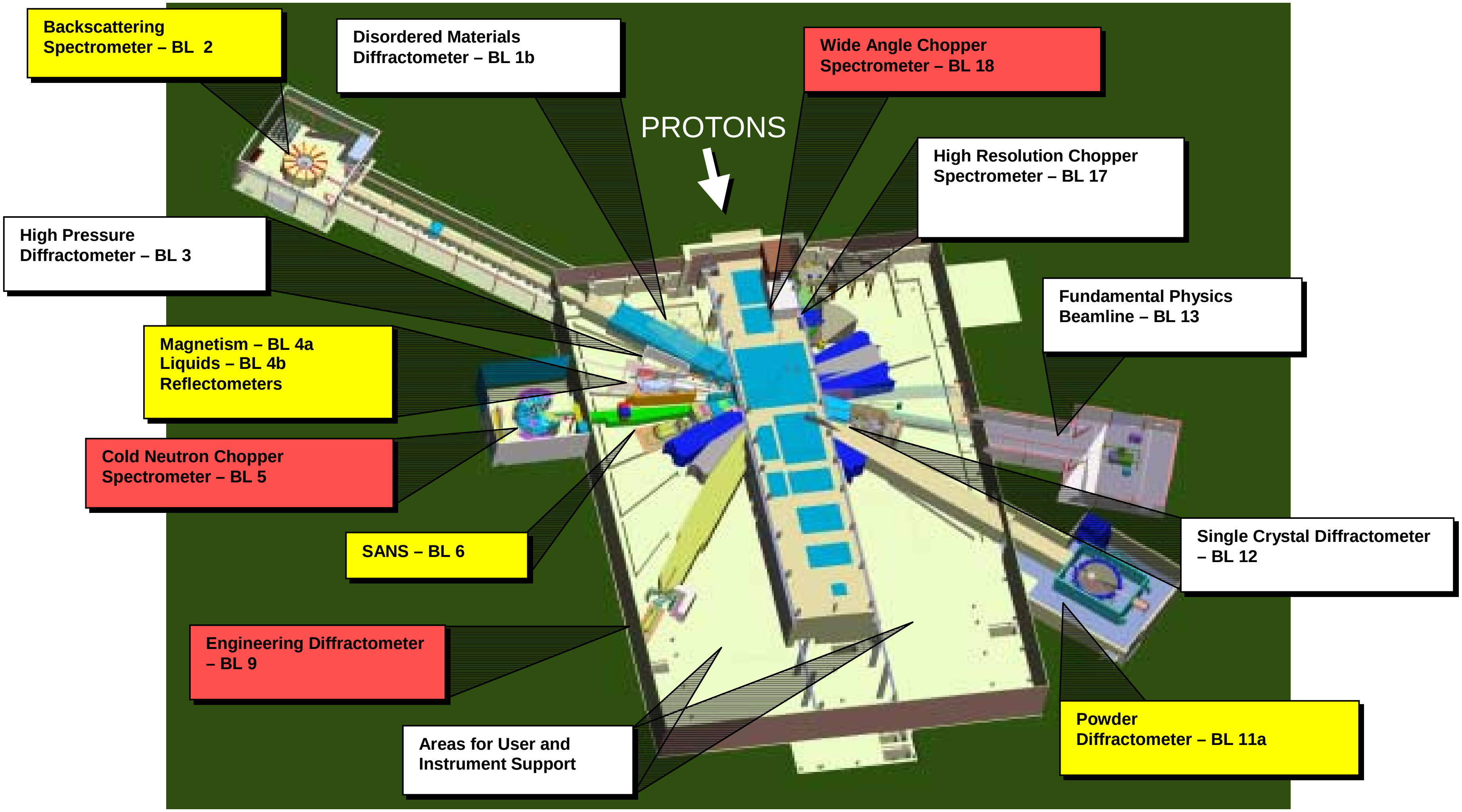



**Fig. 2.2 from the Technical Concepts Document - Prospective Instrument Suite for the SNS-LWTS**

For comparison, the status of instrumentation on the HPTS as of July, 2002 is provided on the next page. Legends in yellow correspond to instruments funded within the SNS project; legends in orange correspond to instruments being funded through Instrument Development Teams (IDT's); legends in white correspond to instruments with IDT's that are actively seeking funding.

## Major Findings

The major findings are found in the section below, entitled "Scientific Input into the SNS Instrumentation Suite"; in Appendices A and B containing workshop reports and final reports of the grant's subawards; and in the separate .pdf file containing the report entitled "Technical Concepts for a Long-Wavelength Target Station for the Spallation Neutron Source."


Backscattering Spectrometer – BL 2
Disordered Materials Diffractometer – BL 1b
Wide Angle Chopper Spectrometer – BL 18
PROTONS
High Resolution Chopper Spectrometer – BL 17
High Pressure Diffractometer – BL 3
Fundamental Physics Beamline – BL 13
Magnetism – BL 4a
Liquids – BL 4b
Reflectometers
Cold Neutron Chopper Spectrometer – BL 5
SANS – BL 6
Single Crystal Diffractometer – BL 12
Engineering Diffractometer – BL 9
Areas for User and Instrument Support
Powder Diffractometer – BL 11a

## Scientific Input into the SNS Instrumentation Suite

**The Spallation Neutron Source**

DOE is funding construction of a major new user facility, at ORNL in Tennessee. Designed to operate at a power level of up to 2 MW - more than an order of magnitude higher than ISIS in the UK, currently the world's most intense pulsed spallation neutron facility - the SNS will provide the research community with neutron beams of unprecedented intensity for a new generation of experimental studies of the structure and dynamics of materials. Both current and prospective users of neutron beams from North America, Europe and Japan have been prominently involved from the beginnings of the SNS project in specifying the performance characteristics for a robust suite of SNS instruments. These instruments will cover a significant range of momentum and energy transfers that will enable new science in a range of disciplines. This involvement is documented below in the section entitled "Linking Science Drivers to SNS Instrumentation".

Pulsed neutron instrumentation employs time-of-flight techniques to determine neutron velocities, and hence wavelengths and energies. Thus the flight path length and source pulse width determine the instrument resolution. The length of the flight path and the time between pulses determines the bandwidth; that is, the range of energy or length scales accessed by a spectrometer or diffractometer. Ideally, that time should match the range of length or time scales of interest in the systems under study, and the pulse duration should be only as long as can be tolerated by the resolution requirements of the science. In general, short wavelength (~0.5-3 Å) neutron instruments employ flight paths of about 10-20 m, require shorter neutron pulses (tens of μsec), and can run at higher frequencies (30-120 Hz). Long wavelength (2-20 Å) neutron instruments tend to employ longer flight paths (20-100 m), work at lower frequencies (10-20 Hz), and can often tolerate longer pulse duration (hundreds of μsec).

The SNS was designed from the outset for operation with two, differently optimized, target stations; the High-Power Target Station (HPTS) and the Long-Wavelength Target Station (LWTS). The decision to adopt the HPTS as the initial target configuration in the DOE-funded project is based on the following facts: (1) the technical demands at a given power level are reduced for more frequent, lower-power pulses, and (2) 60 Hz is well matched to about 50% of the instruments envisioned for the facility. With compromises, such as chopping out

**Why Neutrons are Exceptionally Useful Probes**

*Neutrons are electrically neutral; they penetrate centimeters of most materials, enabling in-situ studies;*

*Neutron cross sections exhibit no regular dependence on atomic number and are similar in magnitude across the periodic table, so they are sensitive to light elements in the presence of heavy ones;*

*Certain large differences in isotopic scattering cross sections (e.g., H/D and $^{6}Li/^{7}Li$) make neutrons especially useful for the study of light atoms in materials;*

*The range of momentum transfers available allows researchers to examine a broad range of length scales (0.1 to $10^{5}$ Å); this capability is important for many different materials and applications;*

*Thermal and cold (long-wavelength) neutrons cover a range of energies sufficient to probe a wide range of atomic or magnetic excitations (1.0 to $10^{-7}$ eV), as well as slow dynamical processes such as polymer chain reptation, on time scales up to $10^{-7}$ seconds;*

*Neutrons have magnetic moments and are sensitive probes of magnetic ordering and excitations;*

*Neutrons can be polarized, allowing separation of the nuclear and magnetic cross sections;*

*The simplicity of the magnetic and nuclear interactions facilitates straightforward interpretation of results.*

unwanted neutron pulses, instruments requiring lower frequencies can be placed on HPTS beamlines with intensity losses corresponding to the fraction of pulses used. However, spatial constraints limit the number of instruments that the HPTS can accommodate. Furthermore, because specialization is the hallmark of high-performance neutron scattering instruments, both the range of science that HPTS instruments facilitate and their optimization to do that science would be compromised by trying to place all instruments on the HPTS. Timely construction of the LWTS will offer a different set of opportunities for optimization for long-wavelength neutron instrumentation; the LWTS makes it possible to closely tailor moderator spectra and pulse shapes to the needs of instruments that operate best at lower frequency or with particular constraints on pulse width.

The distinguished history and bright future for research using neutron beams has been well-documented in a series of studies, listed in the **References.** Major accomplishments by neutron scatterers over the last 30 years show a strong representation from U.S. researchers, many of whom have earned major scientific awards for their work. An examination of recent history reveals that advances in neutron sources and construction of next-generation neutron instruments and associated novel sample environments will certainly lead to new science, including applications that cannot be foreseen today. In the words of the Oakbrook panel: "*2Who would have predicted the central role of neutrons in superconductivity before 1986 or the current wide use of neutrons in polymers?l*" DOE's Birgeneau panel noted that "*It can be generally stated that the availability of high flux neutron sources, with the best possible capability in cold neutron research, will play an important role in giving American industries a competitive edge in future soft materials world markets.*" Similarly, the National Academy of Sciences study on condensed-matter and materials physics speaks of a new era in which the ability to manipulate increasingly complex materials exhibiting multiple length scales and time-dependent phenomena will be more important than ever. The authors note: "*This new capability to span length scales is bringing the world of atoms and molecules closer to the world of our experience, from the mysteries of quantum mechanics, to the mechanical properties of materials, to the self-assembly of biological systems. Many of these problems, which underlie technological innovation and revolution, could not have been addressed on a fundamental basis even a few years ago.*" The scientific cases for the European Spallation Source (ESS) and a second target station at ISIS, as well as the European Round-Table for Neutron Beam Sources make similar points both about enabling new science and engineering and about the increasing impact of neutron sciences on the creation of wealth.

Examples of this new science are plentiful. For example a backscattering spectrometer with 200 neV resolution - 10 times better than is available in the U.S. today - will reveal new information on growth processes and chemical reactions through spectroscopy of rotational tunneling, translation, diffusion, and hindered motion that is complementary to NMR. High intensity is useful for studying smaller or weakly-scattering samples, important for example in determining details of polymer chain conformations in thin films using hydrogen/deuterium contrast. Off-specular reflectivity, providing access to in-plane momentum transfer, will reveal details of two-dimensional structures in magnetic multilayers, on surfaces, and in complex polymer assemblies. It will be possible using high-resolution powder diffraction to determine structural parameters for low-level impurities. For surfactant-stabilized magnetic nanoparticles, atomic and magnetic structure can be determined simultaneously, and the details of surfactant layer growth can be followed in situ. In fundamental physics investigations, improved signal-to-noise, polarized beams, and use of time structure to eliminate systematic errors will provide better insight into the weak interaction.

<u>Linking Science Drivers to SNS Instrumentation.</u>

Since 1996, the headcount of scientists and engineers participating in more than 50 workshops and larger meetings on the science portfolio that SNS supports exceeds 1800. These individuals represent at least 150 different institutions. Through these meetings and other means, the broad neutron user community provides extensive input to the SNS project on the scientific landscape that supports its neutron scattering instruments. This input occurs in several ways:

(a) **Through the SNS Scientific Advisory Committee (1996-2000), the SNS Advisory Board (SAB) and the SNS Experimental Facilities Advisory Committee (EFAC).** Their membership is drawn from the U.S. and international user community and covers the range of disciplines that apply neutron scattering to the study of materials.

**(b) Through Instrument Advisory Teams (IAT's) that develop the case for individual instruments.** After acceptance of an instrument by EFAC for inclusion in the SNS project baseline, an IAT continues to advise SNS instrument scientists on performance requirements.

**(c) Through both comprehensive user workshops and small workshops that focus area by area on the science/instrumentation linkage.** The first of the comprehensive workshops listed below – held in 1996 - recommended desirable performance parameters for a strawman suite of 37 instruments. Approximately one-half of these can take full advantage of source operation at 60 Hz. The remaining instruments – because the science they support requires longer-wavelength neutrons – need operating frequencies of 20 Hz or less. A small fraction of the highest-priority instruments have been accepted by EFAC for the SNS High-Power Target Station. The comprehensive workshops held in 1998 and 2000 built on and refined the instrument characteristics and added new instrument concepts. Applications of pulsed neutron beams other than scattering are also a focus of the neutron user community.

**Comprehensive Workshops.**

1. Workshop on Instrumentation Needs and Performance Metrics for the National Spallation Neutron Source – 10/31 to 11/1/96, Oak Ridge, TN.

2. SNS Neutron Instrumentation Workshop and Oak Ridge Neutron Users Meeting – 11/9 to 11/11/98, Knoxville, TN. [Funded by a grant from NSF-DMR, Lee Magid, University of Tennessee, PI.]

3. Spallation Neutron Source Users Meeting – 5/22 to 5/24/00, Washington, D.C.

4. Two sessions at the March, 1998 "Neutron Science Symposium – Scientific & Industrial Opportunities for the Spallation Neutron Source", held in Washington, D.C., were devoted to presentations on community needs in neutron science.

5. American Conference on Neutron Scattering (ACNS), 6/23 to 6/27/2002, Knoxville, TN.

**Focused Workshops.**

1. Break-out sessions and their chairs at the 1996 comprehensive workshop:
   a) Excitations – Rob Robinson (LANL)
   b) Powder Diffraction – James Jorgensen (ANL)
   c) Single-Crystal Diffraction – Bryan Chakoumakos (ORNL)
   d) Small-Angle Neutron Scattering – Lee Magid (Tennessee)
   e) Reflectometry – Bill Hamilton (ORNL)

2. Focused workshops and their chairs, held in preparation for the 1998 comprehensive workshop:
   a) Magnetism – Jim Rhyne (Missouri)
   b) Advanced Materials for Extreme Environments – C.K. Loong (ANL)
   c) Liquids and Disordered Materials – David Price (ANL) and Brian Annis (ORNL)
   d) Chemistry and Soft-Matter Physics – Frans Trouw (ANL), Grant Smith (Utah), Bruce Hudson (Syracuse), others
   e) Crystallography – James Jorgensen (ANL)
   f) Applied Science and Engineering – Jim Richardson (ANL) and Tom Holden (Chalk River)
   g) Large Scale Structures – Brent Heuser (Illinois) and Lee Magid (Tennessee)

3. Break-out sessions and chairs at the 1998 comprehensive workshop:
   a) Magnetism - Rob Robinson (LANL)
   b) Novel Materials in Extreme Sample Environments - H. Strauss (UC-Berkeley)
   c) Liquids and Disordered Materials - M. Winokur (Wisconsin)
   d) Chemistry - Bruce Hudson (Syracuse)
   e) Crystallography - Bill David (ISIS)
   f) Engineering Materials - Tom Holden (Chalk River)
   g) Large-scale Structures - B. Heuser (Illinois)
   h) Overview of break-out sessions – Henry Glyde (Delaware)

4. Focused workshops held in 2000 associated with the NSF-DMR supported conceptual design study for an SNS Long-Wavelength Target Station (LWTS) are listed below. Most of these groups also held break-out sessions at the 2000 comprehensive workshop.
   a) Soft Matter – Kent Blasie (Penn) and Rob Briber (Maryland)
   b) Magnetic Materials – Collin Broholm (Johns Hopkins) and Dmitri Argyriou (ANL)
   c) Disordered Materials – Henry Glyde (Delaware) and C.K. Loong (ANL)
   d) Crystallography – Angus Wilkinson (Georgia Tech) and James Jorgensen (ANL)
   e) Chemical Spectroscopy and Dynamics – Heloisa Bordallo (ANL) and Kent Blasie (Penn)
   f) Macromolecular Single-Crystal Diffraction at the SNS – Chris Dealwis (Tennessee)

6. International Workshop on Fundamental Physics with Pulsed Neutron Beams – 6/1 to 6/3/2000, Research Triangle Park, NC. Co-Chairs: G. Greene (LANL), M. Snow (Indiana), C. Gould (NC State) and F. Plasil (ORNL).

7. Joint Institute for Neutron Sciences (JINS) Workshop on Applications of Neutron Scattering to Materials Science and Engineering, 10/1 to 10/3/2001, Oak Ridge, TN. 160 participants.

8. JINS Workshop on Neutron Scattering Applied to Structure and Dynamics in Biological Systems, 4/8 to 4/10/2002, Oak Ridge, TN. 100 participants.

9. Breakout sessions at ACNS, 2002:
   a) SNS Cold Neutron Chopper Spectrometer – Paul Sokol (Penn. State)
   b) Engineering Materials – Hahn Choo (Tennessee)
   c) SNS sample environments – Lou Santodonato (SNS)
   d) SNS Magnetism Reflectometer – Suzanne te Velthius (Argonne)
   e) SNS High-Resolution Backscattering Spectrometer – Ken Herwig (SNS)
   f) SNS Single Crystal Diffractometer – Christina Hoffman (SNS)
   g) Data Storage using the NeXus Format – Tom Worlton (Argonne)
   h) Future Relevance of Neutron Reflectometry to Soft-Matter Science – John Ankner (SNS)

### Planning Workshops Focused on the Science Case for Applications Using Cold Neutrons

Members of the U.S. and international user community also provided input to the SNS project at several planning workshops convened by the SNS-LWTS academic consortium (Tennessee, Delaware, Georgia Tech, Indiana, Illinois, Johns Hopkins, Penn, UMass, Harvey Mudd) and by the LWTS target/moderator and instrument groups at ANL. These included:

1. Proposal planning meeting at the University of Delaware, April, 1999.
2. Review of target concept by international experts at ANL, January, 2000.
3. Joint meetings on the first-draft science case by the academic consortium and the target/moderator and instrument groups at ANL, Sept. and Oct., 2000.
4. User workshop at ANL, with approximately 100 participants, to review the LWTS science case, Nov. 20-21, 2000.

**Background Materials.** *[These are found on a CD produced in May, 2001 entitled "Scientific Input into the SNS Instrumentation Suite. The CD was provided to NSF-MPS and NSF-DMR officials. Copies are available on request.]*

Membership lists for the SNS Scientific Advisory Committee, the Experimental Facilities Committee and the SNS & HFIR User Group (SHUG) are available in the Advisory Committee folder.

Membership lists for the Instrument Advisory Teams are available at www.sns.anl.gov and in the Instrument Teams folder. Information is also available there for Instrument Development Teams (IDT's) , groups of scientists with focused needs who acquire non-project funding for instrument design and construction in return for dedicated access for their research programs. IDT's are in place for a Fermi Chopper Spectrometer (ACRS) and for a 10-100 μeV multichopper spectrometer (also called the Cold Neutrons Chopper Spectrometer, CNCS).

Reports of the Experimental Facilities Advisory Committee (originally called the Instrument Oversight Committee) are available in the EFAC-IOC reports folder.

Reports for many of the comprehensive workshops and associated focused workshops are also available in the Workshop Reports folder.

1996 Workshop on Instrumentation Needs and Performance Metrics for the National Spallation Neutron Source (includes break-out sessions): 96user.pdf

1998 SNS Neutron Instrumentation Workshop and Oak Ridge Neutron Users Meeting (includes break-out sessions and separate, focused workshops): 98novuser.pdf and 98xxxxx.pdf. This report is also linked from the NSF Website, www.nsf.gov/mps/divisions/dmr/research/start.htm.

2000 Spallation Neutron Source Users Meeting: 00user.pdf

Reports of the LWTS planning workshops – and some break-out sessions at the May, 2000 users meeting – are available as 00xxxxx.pdf

Agendas for the comprehensive workshops are co-located with the workshop reports. Attendees' institutional affiliations are available in the User Affiliations folder.

The folder called LWTS proposals contains the full proposal on the proposed Long-Wavelength Target Station submitted to NSF-DMR in February, 2001. It also contains the planning grant proposal.

## List of References


F. Seitz and D. Eastman (NRC Major Materials Facilities Committee), *Major Facilities for Materials Research and Related Disciplines,* National Academy Press (1984). (http://www.sc.doe.gov/production/bes/BESAC/major.pdf)

W. Kohn, ed., *Neutron Sources for America's Future*, Report of the Basic Energy Sciences Advisory Committee Panel on Neutron Sources (January 1993). (http://www.sc.doe.gov/production/bes/BESAC/Neutron%20source%20America%20Future.pdf)

D.L. Price and J.J. Rush, Editors, *Neutron Sources and Applications*, Report of a 1992 Review Held at Oak Brook, Illinois for the Basic Energy Sciences Advisory Committee Panel on Neutron Sources (January 1994).

Report of the European Science Foundation Exploratory Workshop, *"Scientific Prospects for Neutron Scattering with Present and Future Sources,"* Autrans, France (1996). (http://www1.psi.ch/www_ensa_hn/ensa/autrans.pdf)

J.L. Finney, U. Steigenberger, A. D. Taylor, C.J. Carlile, and J. Kjems, Editors, *ESS: A Next Generation Neutron Source for Europe, Volume II, The Scientific Case* (March 1997).

R. Birgeneau, G. Aeppli, and T. Russell, panel chairs, *Report of the Basic Energy Sciences Advisory Committee on Neutron Source Facility Upgrades and the Technical Specifications for the Spallation Neutron Source* (March 1998).

Final Report of the SNS Neutron Instrumentation Workshop and Oak Ridge Neutron Users Meeting, Knoxville, TN (http://www.ornl.gov/jins/sns.htm) (November 1998).

D. Richter and T. Springer, *A Twenty Years Forward Look at Neutron Scattering Facilities in the OECD Countries and Russia* (http://www.oecd.org/pdf/M00004000/M00004506.pdf)

The European Round-Table for Neutron Beam Sources (http://www.fys.risoe.dk/fys/ron/neutrons/roundtable/Brochure).

J. Wise (NRC Committee on Developing a Federal Materials Facilities Strategy), *Cooperative Stewardship - Managing the Nation's Multidisciplinary User Facilities for Research with Synchrotron Radiation, Neutrons, and High Magnetic Field*s, National Academy Press (1999). (http://www.nap.edu/books/0309068312/html/index.html)

A second Target Station at ISIS (http://www.isis.rl.ac.uk/targetstation2) - Science Case and Draft of Instrument Suite.

J.J. Rush, presentation to the NRC Committee on Developing a Federal Materials Facilities Strategy (September 1998).

European Neutron Scattering Association, *The ENSA Survey of the European Neutron Scattering Community and European Neutron Facilities* (December 1997). (http://www1.psi.ch/www_ensa_hn/welcome_ensa.html)

1998 Report of the Structural Biology Subcommittee of the Biological and Environmental Research Advisory Committee – DOE-Office of Science, Biological and Environmental Research (http://www.er.doe.gov/production/ober/berac/final697.html).

"Condensed-Matter and Materials Physics: Basic Research for Tomorrow's Technology," p. 7 NAS/NRC Board on Physics and Astronomy, Committee on Condensed-Matter and Materials Physics, National Academy Press, Washington, D.C. (1999).

Report on the Status and Needs of Major Neutron Scattering Facilities and Instruments in the United States. Office of Science and Technology Policy, Interagency Working Group on Neutron Science. (http://www.ostp.gov/html/NeutronIWGReport.pdf)

Report of the Basic Energy Sciences Advisory Committee Subpanel on Neutron Scattering, DOE/SC-0017, February 2000, ("Blume/Rowe report"). (http://www.sc.doe.gov/production/bes/BESAC/neutronrpt.pdf)

Research Infrastructures: Neutron Sources, Edited by Campbell Warden, Reports of the European Roundtable for Neutron Sources, European Commission, 2001.

# APPENDIX A: Reports of the Focused Workshops Supported by the Grant

**Notes on the Contents**

[Workshops held in 2000, with the exception of the workshop on fundamental physics using neutrons, were supported by this grant. The workshop on vibrational spectroscopy using neutrons, held in Feb., 2002, was also supported by this grant.]

1. Soft Matter Working Group – report of their breakout session at the May, 2000 users meeting conveys the results of the April, 2000 workshop as well.

2. Magnetic Materials Working Group – no report of the April, 2000 workshop. Report of their breakout session at a November, 1998 users meeting conveys the sense of the community.

3. Disordered Materials Working Group – report of the April, 2000 workshop.

4. Crystallography Working Group – Their focus is on powder diffraction; report of their May, 2000 workshop is included.

5. Chemical Spectroscopy and Dynamics Working Group – report of the Oct., 2000 workshop. This working group focused on the use of high resolution backscattering and neutron spin echo spectroscopy.

6. Structural Biology Working Group – report of the Dec., 2000 workshop.

7. Vibrational Spectroscopy: formation of an instrument development team for a proposed SNS spectrometer called VISION. Report of their Feb., 2002 workshop is included.

8. Fundamental Physics using Neutrons – report of their June, 2000 workshop.

**SNS Soft Materials Working Group Re-SNS-LWTS**
**[SNS USERS MEETING 5/22-24/00]**

The soft matter working group chaired by J.K. Blasie (U. Pennsylvania) and R.M. Briber (Univ. of Maryland) met on Tuesday 5/23/00 at the SNS Users meeting held in Washington, DC. Presentations were made by Ken Herwig, John Ankner and J.K. Zhao of the SNS project. Ken Herwig discussed the 2 μeV backscattering instrument on the HPTS and a proposed 200 nano(eV) backscattering instrument for the LWTS. John Ankner discussed the liquids reflectometer for the HPTS and possible designs for a broad bandwidth horizontal sample geometry reflectometer and a grazing incidence small angle spectrometer for the LWTS. J.K Zhao discussed the high intensity high precision SANS instrument for the HPTS and a similar SANS instrument for the LWTS.

Agenda

| | |
|---|---|
| 12 noon – 12:15 | R.M. Briber / J.K. Blasie Introduction |
| 12:15 - 12:45 | K. Herwig 2 μeV Spectrometer / 200 nano(eV) Spectrometer |
| 12:45 - 1:15 | J. Ankner Reflectometry and Grazing Incidence |
| 1:15 - 1:45 | J.K. Zhao SANS |
| 1:45 – 4:00 | Discussions: Instruments and Science |
| 4:00 – 5:00 | Reports from Breakout Sessions |

The group spent about 2 hours discussing both the HPTS instruments and the specifications of various new instruments for the LWTS. The most lively discussion centered about the need for (ultra)low Q in SANS measurements. It was decided that that a design goal would be for an initial instrument on the LWTS would be a general purpose SANS instrument with $Q_{min}$=0.0001Å$^{-1}$. A specialized low Q instrument with focusing optics would be lower priority.

A participant list is included at the end of this document.

The overall conclusions reached by the group can be divided into those related to the HPTS and LWTS respectively.

Summary: Soft Matter Working Group

**HPTS**

1.) The working group has strong support for the High Intensity High Power SANS (HIHP-SANS) instrument on the HPTS. The goal should be to get this instrument approved for starting engineering design by the IOC.

2.) The group has concerns that the vertical sample geometry polarized reflectometer is being targeted exclusively at the magnetic materials community. There are a number of people working in other fields who are interested in this machine. The working group proposes that the 2 HPTS reflectometer IATs be combined into one IAT.

**LWTS**

1.) Work should proceed on the preliminary design of a general purpose SANS instrument for the LWTS. This should be one of the 3-4 instruments identified for more detailed study in preparation for the LWTS full proposal to NSF.
Instrument characteristics:
High intensity
$Q_{min}$=0.0001Å$^{-1}$
Large sample chamber able to accommodate a broad range of sample environments, sample holders, etc.

2.) The group had strong support for a broadband reflectometer for the LWTS. This would be an instrument designed to cover a broad range in Q in a single geometry. It would be suitable for studying the kinetics of interfacial reactions, liquid-liquid interfaces, etc.

3.) There was strong interested in the 200 nano(eV) backscattering instrument proposed for the LWTS by Ken Herwig. The group needs to work on building the community and the scientific case for such an instrument. There has not been much inelastic neutron scattering activity in the soft matter community in the US (unlike Europe) due to the lack of instrumentation. This is poised to change with the new instruments at NIST but work needs to be done to develop support for this type of instrument (and similarly a spin echo instrument) at the SNS.

4.) John Ankner presented ideas on a true grazing incidence small angle scattering instrument (GISANS). This would be new type of instrument which uses a grazing incidence incident beam ($\theta < \theta_c$) and measures the scattering from the evanescent wave generated at the surface. It is different from near surface SANS where the beam is confined to the surface by attenuation effects (with $\theta > \theta_c$). This type of experiment has not been pursued for neutron scattering but has become common (but probably not routine) with x-rays at synchrotron sources. The SNS has the potential to allow the development of this technique.

5.) There is a general concern over signal to noise ratios at the SNS (both HPTS and LWTS) due to potentially high backgrounds. This lead to a couple of recommendations:

a.) The proposed slab moderator design for the LWTS has the potential for a large high energy neutron background due to the guides being in direct line of sight of the target. It is recommended that a flux trap target/moderator design be explored for the LWTS with the goal of minimizing the background. Some trade-off of flux would be acceptable in exchange for a significantly lower background.

b.) A Monte Carlo study should be done to assess the effect of including a To chopper *in addition to using curved guides* for the HIHP SANS instrument on the HPTS. Inclusion of a To chopper would result in a loss of intensity of about 15-20% due to the removal of section of guide for insertion of the chopper. This is a reasonable trade-off if there would be a similar or larger decrease in the background intensity.

**Participant List Soft Matter Working Group**

| | Name | Affiliation | e-mail address | Interests |
|---|---|---|---|---|
| 1. | K. Blasie | U. Pennsylvania | jkblasie@sas.upenn.edu | Biomolecular Mat'ls |
| 2. | R. Briber | U. Maryland | rbriber@eng.umd.edu | Polymers |
| 3. | K. Herwig | ORNL | kherwig@anl.gov | Diffusive/Biomolecules |
| 4. | J. Zhao | ORNL | zhaoj@ornl.gov | SANS |
| 5. | B. Hudson | Syracuse U. | bhudson@syr.edu | Biopolymers |
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| 7. | E. Gilbert | ANL | egilbert@anl.gov | |
| 8. | G. Smith | LANL | gsmith@lanl.gov | Surface Sci./LiqXtals |
| 9. | W. Hamilton | ORNL | hamiltonWA@ornl.gov | Surfaces/Complex Fluids |
| 10. | J. Ankner | ORNL-SNS | jankner@anl.gov | Thin Films |
| 11. | M. Pollard | U. Mass. | mpollard@mail.psc.umass.edu | SANS/Polymers |
| 12. | M. Dadmun | U. Tenn. | Dad@utk.edu | Polymers |
| 13. | P. Yuan | ORNL | yuan@phonon.ssd.ornl.gov | Biophysics |
| 14. | M. Foster | U. Akron | foster@polymer.uakron.edu | Polymer/Surface/SANS |
| 15. | M. Kent | Sandia Nat. Lab | mskent@sandia.gov | Polymer/Biopolymer |
| 16. | S. Yun | U. Maryland | augustin@eng.umd.edu | Polymer |
| 17. | S. Choi | U. Maryland | choisw@eng.umd.edu | Polymer |
| 18. | M. Popovici | MURR | popovicim@missouri.edu | Instrumentation |
| 19. | D.L. Ho | U.MD/NIST | derek.ho@NIST.GOV | Polymer |
| 20. | D. Vaknin | Ames Lab | vaknin@ameslab.gov | |
| 21. | P. Butler | ORNL | butlerpd@ornl.gov | Complex Fluids/Flow |
| 22. | U. Perez-Salas | U. Maryland | ursala@eng.umd.edu | |
| 23. | S.H. Chen | MIT | sowhsin@mit.edu | Soft Matter |
| 24. | P. Thiyagarrajan | ANL | thiyaga@anl.gov | Soft Matter |
| 25. | M.Z. Hu | ORNL | i5h@ORNL.GOV | Thin Films/Nano/SA |
| 26. | D.F. Chen | ANL | dfchen@anl.gov | |
| 27. | J. Lal | ANL | jlal@anl.gov | Soft Matter |
| 28. | H.D. Cochran | ORNL | hdc@ornl.gov | Soft Matter |
| 29. | D. Schaeffer | U. Cincinnati | | |

**Magnetism Working Group and Breakout Session**
**Held in Conjunction with the SNS User's Meeting**
**Knoxville, Tennessee**
**7-11 November 1998**
**[compiled by J.F. Ankner]**

**SNS document ES.1.1.8.4.5000.RA.00**

# I. Participants

## I.A. Working Group Members and Activities

| | |
|---|---|
| Jim Rhyne | U. of Missouri (Chair) |
| Ken Andersen | ILL |
| Julie Borchers | NIST |
| Gian Felcher | Argonne National Lab |
| Eric Fullerton | IBM Almaden |
| Bruce Gaulin | McMaster U. |
| David Lind | Florida St. U. |
| Paul Miceli | U. of Missouri |
| Ray Osborn | Argonne National Lab |
| Rob Robinson | Los Alamos National Lab |
| John Tranquada | Brookhaven National Lab |
| John Ankner | SNS Project (Secretary) |

The Working Group met Saturday and Sunday, November 7-8, 1998, at the Hyatt Regency Hotel in Knoxville, Tennessee. After individual introductions and a welcome by the Chairman, Thom Mason, Deputy Director for Science of the SNS, outlined the project and described the instrument selection process. Each group member then gave a 30-minute presentation (with questions) of the scientific needs of their particular sub-field of magnetism. Afterwards, most of the Group members adjourned to Calhoun's Restaurant for an evening of informal discussion, beer, barbecue, and scientist-UT alumni interaction (predominantly inelastic).

Sunday morning, the Group discussed general instrumentation requirements and then broke into two sections, one focusing on thin film studies and the other on magnetic excitations and crystallography. The results of these discussions, transcribed during the afternoon by various Group members, form the backbone of this document.

## I.B. Breakout Session Attendees and Activities

| | |
|---|---|
| Rob Robinson | Los Alamos National Lab (Spokesman and Moderator) |
| Ken Andersen | ILL |
| John Ankner | SNS Project |
| Craig Barnes | U. of Tennessee |
| Dave Belanger | U. of California at Santa Cruz |
| Collin Broholm | Johns Hopkins U. |
| Jacky Chen | Georgia Institute of Technology |
| Kurt Clausen | Risø National Laboratory |
| P. Dai | Oak Ridge National Lab |
| Jack Davidson | Oak Ridge National Lab |
| John Ditusa | Louisiana St. U. |
| Heather Frase | Caltech |
| Garrett Gronroin | Oak Ridge National Lab |
| Hazuki Kawano | Oak Ridge National Lab |
| Mark Meisel | U. of Florida |
| Wouter Montfrouÿ | Oak Ridge National Lab |

| | |
|---|---|
| Stephen Nagler | Oak Ridge National Lab |
| Stephen M. Shapiro | Brookhaven National Lab |
| Liyong Shen | U. of Alabama |
| Alan Thompson | NIST |
| John Tranquada | Brookhaven National Lab |
| Oswald Uwakweh | U. of Cincinnati |
| Barry Wells | U. of Connecticut |
| Guangyong Xu | Johns Hopkins U. |
| Mohana Yethiraj | Oak Ridge National Lab |
| Jerel Zarestky | Ames Laboratory |

The Breakout Session convened Tuesday morning, November 10, at the Hyatt. The Moderator charged the attendees with formulating scientific instrument specifications and participants were given a preliminary draft of this document as a starting point. During lunch, Steve Nagler described the cold-neutron triple-axis instrument that will be part of the HFIR upgrade and Alan Thompson presented the latest results of $^3$He polarizer development at NIST. After vigorous discussion, the attendees agreed on the prioritized list presented below in the Instrument Requirements section. The Moderator prepared viewgraphs describing these instruments and presented them to the full meeting. Rob Robinson deserves particular commendation for doing a great job as moderator and spokesman without much advance warning.

## II. Scientific Interests

### II.A. Magnetic Thin Films

- Capability to study smaller samples aids in the study of all magnetic films. Reduces problems of uniformity during deposition. Increases the universe of samples that can be studied.
- Measuring long correlation lengths, such as fluxoid lattices in superconductors or magnetic domains. Develop polarized-beam off-specular data collection and data analysis techniques.
- Magnetic dot lattices: what is the collective response of arrays, study intrinsic vs. statistical properties. Non-uniformity of dots is a problem.
- Physics in confined geometries is intrinsically different than in bulk. Can study reduced dimensionality and the effects of pressure.
- Relaxation processes in magnetic films – response to 2 ns excitation of great technological interest. Exploit capability of pulsed source to measure reversible, time-dependent relaxation phenomena.
- Inelastic scattering from thin films – how thick is thick enough? Study effects of dimensionality on magnetic excitations.
- Study structural vs. magnetic roughness in both bulk and thin-film geometries. What is the correlation between structural and magnetic properties?
- Understanding the behavior and properties of small magnetic clusters is increasingly important as the size of magnetic devices decreases. Conventional microscopies lack the resolution to study the structure of, e.g. 20 nm clusters in two dimensions.
- Nanoparticles, spin glasses, amorphous, and polycrystalline films – clusters.

- Investigate special properties of surfaces – 5-100 nm length scales are characteristic of many magnetic problems.

**II.B. Magnetic Excitations**

- Low-temperature phase transitions
- Disordered and frustrated ground states
- Quasi-elastic and low-energy excitations
- Short-range magnetic ordering and correlations
- Crystal-field excitations
- Spin fluctuations – correlated electron systems
- Magnetic densities of states
- Coherent excitations of all sorts
- Molecular magnets
- Magnetism as a probe of superconductivity
- Spin waves in ordered and disordered materials
- Quantum tunneling and magnetization
- Amorphous magnets
- Low-dimensional magnetic systems
- Inelastic scattering in films, multilayers, and nanoparticles
- Quantum critical points

**II.C. Magnetic Diffraction**

- Determination of magnetic structure and/or crystal structure in commensurate or incommensurate magnetic order in exotic structures (e.g., rare earths, actinides, and novel superconductors)
- Diffraction from magnetic superlattices, which involves both nuclear and magnetic structure determination. The useful data will come from following a specific trajectory in *Q*-space, for example a line though a principal magnetic reflection and its satellites along the growth axis. Particularly in the case of ferromagnetic materials, a full polarized beam capability is essential.
- Determination of form factors of magnetic ions in thin films or exotic local symmetries.
- Critical behavior of new universality classes
- Novel electronic materials
- Novel ground states and phase diagrams
- Domain physics
- Magnetism as a probe of superconductivity
- Magnetic short-range order
- Spin-lattice coupling
- Organic conductors
- Field-induced magnetism
- Magnetization density maps
- Low-moment magnetism – down to 0.01 $\mu_B$
- Quantum critical points

## III. Instrumental Requirements

The availability of efficient broad-band polarizers for large, divergent neutron beams would make possible the use of polarized-beam techniques on a wide range of neutron instruments. Coupled with the flux enhancements at the SNS, the potential exists for a revolution in the study of magnetic materials. Recent developments at the Institut Laue-Langevin and other laboratories utilizing spin-polarized $^3$He show great promise. The elimination of field gradients caused by stray magnetic fields is essential for the use of $^3$He polarizers. Consideration should therefore be taken to eliminate sources of stray magnetic fields on all instruments that could potentially benefit from the addition of $^3$He polarization capability. Ferrous metals, particularly steel, are the primary source of these stray fields. The presence of stray fields is also a consideration in the safe use of high-field magnets.

## III.A. High-Priority Instruments

### III.A.1. Polarized-Beam Reflectometer

The range of thin-film problems addressed by neutron reflectivity, the inconvenience of installing and de-installing polarizing elements, and the necessity for elimination of stray fields argue strongly for construction of a dedicated polarized-beam instrument. Polarization of a narrow beam such as is used in reflectometry is easily accomplished using supermirrors. For specular reflectivity experiments, supermirrors can likewise be used for exit-beam polarization analysis. However, the growing interest in problems addressed by off-specular scattering, such as the characterization of domains and magnetic dots, can only be served by the development of divergent-beam analyzers such as $^3$He. The small dimensions of many thin-film magnetic structures would be well-served by the bandwidth available at a low-repetition-rate (20 or 30 Hz) second target station. However, the utility of a polarized-beam instrument would be only marginally impacted if located in a 60-Hz hall. In either case, the instrument should view a cryogenic coupled moderator. In order to measure crystalline diffraction from, e.g. artificial multilayers and to employ the highest-field cryomagnets, a horizontal scattering plane (one with the sample surface perpendicular to the floor) is desirable.

Salient Features:

- Horizontal scattering plane with detector movable $0 \le 2\theta \le 90^\circ$
- $R_{\min} \le 10^{-8}$ in polarized mode
- $0.01 < \delta Q / Q < 0.10$
- No magnetic components
- Able to resolve film thickness $10\ \text{Å} \le d \le 10{,}000\ \text{Å}$

### III.A.2. 100 $\mu$eV Spectrometer

The magnetism community will require an inverse geometry spectrometer with modest energy resolution (100 $\mu$eV at the elastic position) for quasi-elastic and low energy excitations with a relatively wide dynamic range (0.2 meV < $\delta E$ < 20 meV). This instrument will address scientific issues associated with a range of magnetic and superconducting energy scales of interest to a host of magnetic materials. In particular it will be important to problems involving low-temperature phase transitions and disordered ground states. An important feature of this instrument is that it be designed to provide continuous angular coverage.

Serious consideration should be given to providing flexibility of the secondary spectrometer configuration, for example allowing variable $k_f$ for each analyzer-detector arm. Measurements of $S(Q)$ within the quasi-static approximation should also be possible within this configuration for single crystal critical scattering and magnetic diffuse scattering.

The instrument should be designed to allow for efficient polarization analysis through the use of either supermirror or $^3$He polarizers. As the areas of interest include mainly low-temperature phenomena, a complete suite of cryogenic and magnet cryostat sample environments are essential. For single-crystal applications the capability to orient the sample cryostat in the beam is also essential.

This instrument can operate efficiently at 60 Hz, although the dynamic range in neutron energy gain would be improved with a lower repetition rate such as 20 or 30 Hz. This instrument should view a cryogenic moderator.

Salient Features:

- $-4\ \text{meV} \le E \le 20\ \text{meV}$
- $\delta E = 100\ \mu\text{eV}$
- $0.1\ \text{Å}^{-1} \le Q \le 4.0\ \text{Å}^{-1}$
- $\delta Q \le 0.02\ \text{Å}^{-1}$
- Polarized incident beam option
- No magnetic components
- High-field sample environment
- Continuous $Q$ coverage
- >100× intensity of equivalent $10\ \mu\text{eV}$ machine

## III.A.3. High-Resolution Direct-Geometry Spectrometer

We believe there is a need for two direct-geometry inelastic spectrometers, both of which could be used for powder or single-crystal experiments. Both will use incident energies between 15 and 1000 meV. One instrument will have higher resolution ($\delta E_i/E_i$ ~ 1%) and will be optimized for polycrystalline problems including crystal-field excitations, some spin-fluctuation problems and phonon densities of states. It should cover energy transfers between 1 and 500 meV, an angular range between 2 and 140 degrees and collect all of the solid angle up to a $30^\circ$ scattering angle. Magnetic materials should be avoided in spectrometer construction (out to a radius of 2 m).

The instrument designers should work with detector manufacturers to get square cross-section PSDs of sufficient length (1 m or more). It is important that both instruments can change/remove Fermi choppers in an automatic manner, without moving shielding or using the crane.

Salient Features:

- $\delta E_i / E_i = 0.01$
- $15\,\text{meV} \leq E_i \leq 1000\,\text{meV}$
- $1\,\text{meV} \leq E \leq 500\,\text{meV}$
- $2^\circ \leq \phi \leq 140^\circ$
- Full coverage for $\phi \leq 30^\circ$
- $\delta Q \leq 0.05\,\text{Å}^{-1}$
- Position-sensitive detectors used everywhere

## III.B. Important Instruments for Magnetism

### III.B.1. High-Intensity Direct-Geometry Spectrometer

The other direct-geometry instrument should be optimized for magnetic scattering from single crystals and can benefit from poorer resolution (~5%) and a corresponding increase in intensity. It should use the same incident energy range (15 – 1000 meV), and might be placed on a coupled or partially-coupled ambient water moderator. See the description above of the high-resolution direct geometry instrument for common features. It will also be the spectrometer of choice for inelastic scattering using the highest (pulsed) magnetic fields (up to 40 T). It should be designed from the outset to accommodate $^3$He polarizers on both incident and scattered flight paths. Magnetic materials should be avoided throughout the spectrometer. This machine will likely have the higher scientific impact.

Salient Features:

- $\delta E_i / E_i = 0.05$
- $15\,\text{meV} \leq E_i \leq 1000\,\text{meV}$
- $2\,\text{meV} \leq E \leq 500\,\text{meV}$
- Full coverage for $\phi \leq 30^\circ$
- $\delta Q \leq 0.1\,\text{Å}^{-1}$
- Position-sensitive detectors used everywhere
- No magnetic components
- >25× Intensity of the 1% machine

### III.B.2. 10 $\mu$eV Spectrometer

The magnetism community could also benefit from a high-resolution inverse-geometry spectrometer.

Salient Features:

- $-1.0\,\text{meV} \leq E \leq 2.0\,\text{meV}$
- $0.1\,\text{Å}^{-1} \leq Q \leq 2.5\,\text{Å}^{-1}$
- $\delta E \approx 10\,\mu\text{eV}$
- $\delta Q \leq 0.02\,\text{Å}^{-1}$
- Continuous $Q$ coverage

## III.C. Instruments Having an Impact on Magnetism

### III.C.1. Single-Crystal Diffractometers

Salient Features:

- Very low and well-characterized background
- No magnetic materials used near beam
- Capability to measure small-moment systems: $\mu \geq 0.01\,\mu_B$
- $0 < Q \leq 7$ Å$^{-1}$
- This instrument should optimally view a cold moderator

In addition to the above instrument, it is highly desirable to have available a second instrument optimized for very high intensity and lower resolution ($\delta d/d$ ~ 1-2%) for quick surveys of magnetic structures or for determination of the temperature-dependence of order parameters. The instrument should also be capable of accommodating extreme environments, particularly high magnetic fields, and high pressures at low temperature. This instrument should optimally view a lower repetition-rate target and view a coupled cryogenic moderator. The experience of LANSCE using a similar instrument should provide guidance for the design of this instrument. The instrument designer should explicitly consider making it magnetically clean (minimal use of ferrous metals).

### III.C.2. High-Intensity Powder Diffractometer

A general-purpose powder diffractometer for structural determination is essential. The length-scale of interest to the magnetism community is $d$ = 2-20 Å. Polarized-beam capability is important. In particular, a detector bank in the forward direction should be equipped for polarization analysis using polarized $^3$He. This requires the elimination of magnetic materials from the instrument construction in the volume including the sample and the forward angle detectors. The ability to solve magnetic structures in conjunction with nuclear structural determination is a very powerful technique which can be realized in this way. The instrument can operate efficiently at 60 Hz.

Salient Features:

- $2$ Å $\leq d \leq 20$ Å
- Polarized incident-beam option
- Polarization analysis using $^3$He in forward direction
- No magnetic materials
- Backscattering detector banks for simultaneous structure determination

### III.C.3. SANS

Ensure that usage of $^3$He polarizer is not hindered by design of incident- and exit-beam flight paths. It is essential for both SANS and reflectometry that the instruments be located on low-background beam lines.

Salient Features:

- Accommodate $^3$He in final flight path and supermirror polarizer in incident
- Accommodate sample magnets
- No magnetic materials

## IV. Ancillary Equipment

- Horizontal and vertical superconducting magnets
- Full complement of cryostats and dilution refrigerators
- Broad temperature range closed-cycle refrigerator (10-500 K)

## V. Areas for Research and Development

- $^{3}He$ polarizers
- Investigate the possibility of multiplexing instruments on 60 Hz source by redirecting beams using supermirrors. By such means, one could, e.g. feed as many as three distinct instruments from a single polarized incident beam.
- Full 3D polarization analysis capability (polarimetry)
- Investigate possibility of performing grazing-angle diffraction at a pulsed source. Survey usage of ILL EVA instrument for scientific relevance.
- Pulsed high-field magnets. View LANSCE initiative as prototype and build on any successes.
- Consider desirability and difficulty of in-situ MBE capability. View NIST efforts to gauge value.
- Evaluate feasibility of grazing-incidence SANS and possibility of testing prototype instruments at existing sources
- Improve chopper designs: higher frequencies, new materials, bearings, electronic choppers
- Evaluate prospects for thermal spin-echo machine
- Polarizing supermirror guides
- Investigate 2 T flux-gated permanent magnet
- Investigate replacing Fermi chopper by focusing monochromator

# REPORT ON
# WORKSHOP ON
# DISORDERED MATERIALS
# LONG WAVELENGTH TARGET STATION
# SPALLATION NEUTRON SOURCE
University of Delaware – April 28-29, 2000

Table of Contents



## PREAMBLE

Below is a report on the Workshop on “Liquids, Glasses and Disordered Materials” held at the University of Delaware, April 28-29, 2000. The workshop was one of four topical workshops held prior to the Spallation Neutron Source (SNS) Users Meeting held in Washington, DC May 22-24, 2000. The workshop was organized jointly by Chun Loong, Argonne National Laboratory and Henry R. Glyde, University of Delaware.

The purpose of the topical workshop was to identify the scientific challenges in the field of Disordered Materials and the opportunities to address these challenges opened by the Long Wavelength Target Station (LWTS) and its associated neutron scattering instruments. A second purpose was to introduce the LWTS and instruments to several major players in the field who do not normally use neutron scattering.

The report consists of (1) The Workshop Program, (2) A list of participants, (3) The Scientific Case for the LWTS, i.e. for structure determination at long wavelength and for determination of low energy excitations in the field of disordered materials and (4) A first cut at translating the scientific case into specific instrumentation needs.

## 1. PROGRAM

**WORKSHOP ON**
**DISORDERED MATERIALS**
**LONG WAVELENGTH TARGET STATION**
**SPALLATION NEUTRON SOURCE**
**University of Delaware — April 28-29, 2000**

***Friday, April 28, 2000***

| | | |
|---|---|---|
| 8:15 | | BREAKFAST, Rm. 206 Trabant Center |
| 9:00 | Henry Glyde | Welcome, Introduction and Purpose of Workshop |
| 9:10 | Lee Magid | SNS-LWTS – Developing the LWTS science case and the full proposal |
| 9:25 | Guebre Tessema | LWTS – Role of National Science Foundation |
| 9:40 | Javier Bermejo | Dynamics of structurally disordered matter: Challenges for next-generation cold neutron instrumentation. |
| 10:10 | Don Kearley | Cold neutrons and numerical methods. |
| 10:40 | | COFFEE |
| 11:00 | Austen Angell | The amorphous state equivalent of crystallization: new glass types by first order transition from liquids and crystals. |
| 11:30 | Dennis Klug | Neutron scattering studies of the structure and dynamics of amorphous ice and related materials. |
| 12:00 | | LUNCH – Rm. 206 Trabant Center |
| 1:30 | Jack Carpenter<br>Ken Herwig | The LWTS and Instrument Selection and Design |
| 2:30 | Herbert Strauss | Neutron spectroscopy of hydrogen gas dissolved in ice. |
| 3:00 | Susan Kauzlarich | Synthesis and Characterization of Group IV Semiconductor Nanoclusters |
| 3:30 | Shenda Baker | Examination of polymer dynamics under shear flow by neutron reflectivity. |
| 4:30 | | Discussion – New Netherland Rm/ Embassy Suites Hotel (Jack Carpenter, Ken Herwig, Chun Loong, Paul Sokol, Herbert Strauss) |
| 5:45 | | RECEPTION – Atrium/Embassy Suites Hotel |
| 7:00 | | DINNER – Fort Casmir Rm/Embassy Suites Hotel |

***Saturday, April 29, 2000***

| | | |
|---|---|---|
| 8:15 | | BREAKFAST – Rm. 206 Trabant Center |
| 9:00 | Marie-Louise Saboungi | Neutrons and soft matter: Lithium conducting polymers. |
| 9:30 | Lennox Iton | Molecules in zeolites. |
| 10:00 | Henry Glyde | Disordered quantum systems. |
| 10:30 | | COFFEE |
| 11:00 | | Discussion and Wrap-up (David Price, Michael Klein, Chun Loong, Ken Herwig and Henry Glyde) |
| 12:30 | | LUNCH and CLOSING REMARKS |

## 2. LWTS WORKSHOP PARTICIPANTS

Dr. C.A. Angell
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## 3. THE SCIENCE CASE: SCIENTIFIC OPPORTUNITIES OPENED BY LWTS

Below we list scientific opportunities and challenges identified at the workshop that can be addressed at the LWTS and the associated neutron scattering instruments. These are listed under six topics. The topics have no specific role or meaning and are simply convenient categories under which the points made at the workshop can be collected.

### A. Liquids and Glasses

**i)** A broad challenge is to reveal the nature of liquids and glasses by determining their excitation energies with high precision over a wide energy ($\omega$) range. This demands high energy resolution

$$\delta\omega \leq 5\text{-}10\ \mu\text{eV over a range } 0 \leq \omega \leq 10\ \text{meV}$$

The goal is to understand the dynamics and thermodynamics of these systems based on precise knowledge of excitation energies. Systematic studies as function of bulk variables such as pressure and composition are needed. This requires regular access to instrumentation and high beam intensity. Some specific issues are: understanding "Boson" peaks in $C_V$, shear modulus in liquids at higher Q values, transitions from free rotation to glassy regime.

**ii)** It was found important to explore dynamics to higher Q values ($Q \geq 2.2\ \Delta^{-1}$). Aim is to cover continuously the Q region in which collective excitations are observed ($Q \leq 2\ \Delta^{-1}$ typically) to higher Q values (up to $Q \cong 5\text{-}10\ \Delta^{-1}$) where single atom (molecule) excitations are observed. Most existing "time of flight" instruments go up to $Q \cong 2\ \Delta^{-1}$ only. Essentially, want to cover a wide Q range of the dynamics. Also seek good Q resolution ($\Delta Q \leq 0.03\ \Delta^{-1}$ mentioned).

**iii)** It would be a great advantage to separate coherent from incoherent scattering. Essentially, in many materials (systems) there is large incoherent scattering which masks the coherent structure and dynamics. The classic example is liquid $H_2$. Need spin-polarized beam and instrument for opportunities noted above in i) and ii) above as for magnetic systems.

**iv)** Annealing of glasses: Goal is to do "real time" studies on structure and dynamics of glasses as they anneal and age. Essentially, need high beam intensity, rapid data collection for this.

**B. Chemical Reactions, Catalysis**

**i)** One aim is to study diffusion and tunneling directly as a means of investigating chemical reactions.

**ii)** Other topics are real time studies of composition change (reactions in progress) as a function of environment and impurities, surfaces and other agents.

**iii)** Investigate structure and dynamics of molecules, systems and impurities on surfaces.

**iv)** Ability to study small samples and small changes in composition is a great advantage here.

**C. Membranes, Proteins**

This topic overlaps with the “soft materials and polymers” group but is of clear interest to several people at the present workshop. Topics identified were:

**i)** Structure of proteins, membranes and macromolecules.

**ii)** Issue of concentration of constituents, currently need to push this up to 10% to get a signal.

**iii)** Isotopic substitution of H/D to reveal role of specific components of large molecules.

**iv)** Study “things” on membranes, e.g. head group changes, impurities, water.

**v)** Dynamics of lipids and proteins, time scales of motion.

These topics need:

— diffractometer for large length scale structures, high intensity
— low energy, high energy resolution spectrometers
— high beam intensity for small samples, small changes in samples
— reflectometry.

**D. Nanostructures**

The important role of neutrons in characterizing the structure, composition, dimensionality and size of nanostructured materials was discussed. This Included replicated materials, powders, structure of porous (especially nanoporous) media and other absorbing and disordering media. Needed here is high resolution SANS at low Q. Given the perceived importance of nanostrcutures, this could be an important user field.

**E. Systems in Porous Media**

The properties of liquids, classical and quantum liquids, in porous media is a field of great current interest. Of special interest is the impact of disorder and confinement on the characteristic excitations and phases of liquid or crystalline systems. For example are there “Bose Glass” phases or new excitations at low energy that destroy order (e.g. superfluidity) in the presence of disorder? Accurate measurement of the change in the structure and of the excitations from the bulk/uniform case to the confined/ disordered case is needed.

Needed here is high Q resolution SANS and high energy resolution spectrometers. High beam intensity to get high statistical precision is also a great advantage when searching for small, subtle changes introduced by disorder.

### F. Films and Substrates

A goal is to measure surface roughness and characterize other properties of surfaces. The properties of impurities and films on these surfaces is a topic of much interest. Properties include structure and phases of films, growth and aging action on surfaces, excitations in the films (3D and 2D), role of impurities in structure and dynamics, surface diffusion, mechanical and other properties.

The same instrumentation as needed for (E) applies here. Of special interest is low Q (L = 40, 60 m) SANS with large Q range and low energy, high energy resolution spectrometers. Capability to study smaller samples and lower impurity concentration is important (requires high intensity).

## 4. INSTRUMENTATION REQUIREMENTS

Some specific comments on instruments by the scientific speakers and participants were:

**i)** Desire for polarized neutrons and polarization analysis routinely to do liquid dynamics separating coherent from incoherent components.

**ii)** Reflectivity for study of surfaces is most important – also with polarized neutrons.

**iii)** A goal is to measure low energy excitations of large systems (e.g. collective excitations in membranes). Need high energy resolution over a wide energy range. Need intensity at low Q.

**iv)** In addition to iii) above, want to determine excitations over a wide Q range, in the range $2 \leq Q \leq 4\ \Delta^{-1}$ as well as lower Q as can do on MARI. However, want $\delta\omega \leq 10\ \mu$ eV. Measure dispersion beyond first Brillouuin zone, density of states $g(\omega)$ at higher Q to average out incoherent effects, to separate different motions.

**v)** Quasi elastic scattering is important – diffusion, tunneling.

**vi)** Want to go to smaller samples, observe impact of small changes in composition in samples. This requires high beam intensity, high statistical precision.

**vii)** Real time studies of systems.

## 5. THE CASE FOR THE LWTS: GENERAL COMMENTS

*— **based on discussions following the workshop and visits to ISIS.***

**A)** In the "Blue Book" (RL-77-064/C) "A pulsed neutron facility for Condensed Matter" edited by L.W.C. Hobbs, G.H. Rees and G.C. Sterling, (June 1977), there is no mention of low energy neutrons. The "Blue Book" is the scientific case document for ISIS but it does not include any discussion of low energy neutrons to study low energy excitations or large structures. That was not foreseen at the time but low energy neutrons have become a major part if not the major part of the program (e.g. IRIS, OSIRIS). Similarly, for the LWTS it is very difficult to look ahead 20 years and articulate precisely the scientific opportunities opened by the LWTS. However, there are clear opportunities now (five years ahead of the LWTS) and the trend is rapidly and clearly in that direction — both in science and in source and instrument capability. A case for the LWTS can be made on the broad trend.

**B)** A case for the LWTS can be made on the total number and variety of instruments. That is, SNS will be the major neutron facility in the USA for some time. A major facility, such as ILL, has approximately 45 instruments. The second target makes this number of instruments possible. The second target particularly makes it possible to build a wide spectrum of instruments covering a wide energy range that can be optimized to take advantage of each target. The HPTS and LPTS enormously improve the instrument capability of the whole facility in variety and performance of each instrument.

**C)** A case for the LWTS can be made on "ownership" of instruments by the scientific community. That is, the NSF funded portion could be regarded as "owned" by the University community. They would have to and want to be directly involved in developing this suite of instruments to its optimum level and take responsibility for its performance. This would be a major step in getting the community directly involved in instrument development and "ownership" in the neutron field as has been achieved in a stepwise way at synchrotron sources.

**D)** Affiliate Institutions. There could be a set of affiliate universities who are affiliated with JINS (UT) and in this way supportive of the LWTS proposal. That is, the proposal comes from JINS or UT but there are "affiliated" universities to JINS that endorse the proposal and add some national representation to the proposal.

**E)** There could be a large number of "mini" biosketches of scientists across the nation who support the SNS attached to the proposal. Each "bio" could in a line or two be connected to a field or instrument. This would demonstrate support in the scientific community and show that it comes from the community.

**F)** The LWTS proposal does not compete with awards to individuals.

- it is a major facility in Materials Science like a telescope or particle physics facility.
- NSF seeks new funds for this CMMS facility as it does for other major facilities. Funds do not come out of existing programs.
- These new funds provide instruments. If the LWTS were not funded as a single new major facility, groups would seek funds for individual instruments. Funded in this way, the instruments would

come out of existing programs.

**G)** Corporations — would some presence in the proposal improve funding prospects?

**Wavelength Powder Diffraction Workshop"**
**held at Argonne National Laboratory, May 12, 2000**
J. D. Jorgensen and A. P. Wilkinson, organizers

The day began with an overview of anticipated LWTS performance from J. W. Richardson. This was followed by a report on the powder diffraction instrumentation for the HPTS from J. Hodges. Ken Anderson from ISIS then provided a useful summary of his experiences with long-wavelength powder diffraction on the OSIRIS instrument at ISIS. Presentations covering some of the scientific opportunities in solid-state chemistry, molecular materials, mesoporous solids, nanoparticulate materials, in-situ catalytic studies, microporous materials, biomolecules and magnetism were made by K. Poepelmeier, P. W. Stephens, A. Stein, R. Whetten, J. Turner, B. Toby, R. VonDreele and D. Argyriou respectively. They were followed by a lively group discussion of scientific priorities and instrument characteristics.

In general, the attendees believed that there were considerable opportunities in the areas of complex highly crystalline materials and materials displaying order on a few nanometer length scale. The possibility of studying organic materials in general and protein structures in particular by powder neutron diffraction received considerable discussion. While the examination of protein structure by powder diffraction methods has recently been demonstrated to be highly effective for certain classes of problem, it is probably too early to tell if powder neutron diffraction could have a big scientific impact on our understanding of protein structure.

The various scientific opportunities broke down into three distinct groups. The successful examination of ordered arrays of nanoparticles and ordered mesoposous solids, such as MCM-41, requires an instrument capable of accessing d-spacings of up to 100 Å, but there is in general no need for information below 2 Å as the mesoporous materials are very poorly ordered on atomic length scales and for the nanoparticulate materials the atomic length scale structure is simple and not readily related to the ordering seen on a several nanometer length scale. For complex large unit cell crystalline materials such as zeolites and many recently developed functional metal oxides it is important to get high resolution data over a wide d-spacing range. The d-spacing range 0.3 – 40 Å was suggested as an appropriate target for a very versatile high-resolution powder diffraction instrument on the LWTS. The move to ever more complex materials is likely to produce considerable demand for this type of instrument. The low repetition rate of the LWTS is also ideally suited to building an ultra high-resolution instrument for examining complex materials over a narrower d-spacing range. Such a long flight path instrument would offer the scattering contrast of a neutron experiment along with resolution that is currently only achieved on synchrotron powder diffractometers.

Ken Andersen's presentation on his experience at ISIS made it very clear that frame overlap problems severely restrict the d-spacing range that can be cleanly obtained using a high repetition rate source of neutrons. Some progress towards the desired instrument characteristics might be achieve at the HPTS with a chopper inside the target biological shield, but we would be much better off trying to build an instrument covering a wide d-spacing range on the LWTS. Additionally, Jim Richardson's presentation indicated that for a given path length, the probable moderator characteristics at LWTS would give us better resolution than could be achieved on the HPTS.

The scientific opportunities discussed seemed to dictate three different instruments: 1) a low resolution instrument spanning the d-spacing range 2 – 100 Å, this idea should probably be developed along with the small angle scattering community, 2) a high resolution medium flight path instrument capable of covering the d-spacing range 0.3 – 40 Å, this would also probably be of interest to the magnetic materials community, and 3) an ultra high-resolution long flight path instrument perhaps covering the d-spacing range 0.3 – 4 Å. These instruments distinguish themselves from what is possible on the HPTS by offering access to an extended d-spacing range and superior resolution. Where count rate it as at a premium and d-spacing range/resolution are secondary considerations, for example during parametric studies, the HPTS instruments will clearly be superior.

## Workshop on Chemical Spectroscopy, Protein Folding Dynamics and Polymer Dynamics

### The 200 neV back-scattering instrument proposed for the Long Wavelength Target Station: Systematic studies over a large dynamic range

**Summary statement**. The proposed 200 neV spectrometer offers a remarkable Q-range $0.05\ Å^{-1} \le Q \le 1.2\ Å^{-1}$, with a high Q resolution of $0.004\ Å^{-1} < \Delta Q < 0.01\ Å^{-1}$ and excellent dynamical range of $-420\ \mu eV < \omega < 420\ \mu eV$. This spectrometer will in conjunction with related instrumentation at the LWTS provide unprecedented opportunities in the areas of chemical and biomolecular dynamics. The complete set of SNS spectrometers, including the proposed LWTS neV instrument, will open in the United States the opportunity to the study of atomic and molecular diffusion in biological systems such as proteins and lipids.

Although much has been learned about the function of biomolecules from a detailed knowledge of their structure, information on the wide variety of motions exhibited by these molecules is also required if we are to fully understand their operation as molecular machines. As well as understanding the principles that lead from sequence to structure ('the protein folding problem'), we also need to understand those principles that lead from structure to dynamics and function. Moreover, the study of macromolecular dynamics provides us with a rather stringent test of the intermolecular forces, which are the basis for understanding protein stability and rationalizing protein design.

Experimental studies of macromolecular dynamics normally utilize optical techniques (photon correlation spectroscopy, fluorescence correlation and photobleaching recovery techniques and Brillouin light scattering) which cover relatively long time scales as compared with the relevant neutron spectroscopic methods of quasi-elastic scattering and spin-echo spectroscopy. Multidimensional NMR has also been used in the elucidation of slow local incoherent dynamical motions and it does not provide information on long range correlation.

Neutron scattering spectroscopies on the other hand cover a much wider dynamic range from, for example, the study of fast molecular diffusion in porous media, segment dynamics to reptation, membrane fluctuations, as well as individual dynamics of molecules tethered to a surface. Also neutron spectroscopy offers the unique possibility to investigate membrane composition effects on protein dynamics. The methodologies therefore are complementary rather than competitive. There is enormous potential for new insights into biomolecular dynamics from the use of quasi-elastic and inelastic neutron scattering at pulsed neutron sources. The time of flight methods used at such sources offer inherent advantages, including potentially greater Q resolution, a larger dynamic range, access to the lowest energy transfers, and high-energy resolution. A particularly important new direction consists of accessing slow (~$10^{-7}$ s) dynamics with the use of very cold neutrons.

Among the many important problems of macromolecular dynamics, that we expect to be able to address at this new facility, are the motions of parts of the lipid molecules (e.g. the head groups) which can be distinguished by selective deuteration, and the associated dynamical processes with different time scales (e.g. the chain dynamics from the lateral diffusion), which will be distinguishable by the use of different spectrometers. Collective and molecular motional processes may be evaluated by the combination of spin-echo and backscattering instrument respectively. In-plane and out-of-plane motional processes may be separated by variation of the scattering angle and appropriately orientational samples. Quasielastic measurements of highly oriented bilayer stacks at different hydration levels answer such questions as why these stacks do not swell indefinitely under maximum hydration, providing insights to the basic intermolecular forces acting between membranes.

Perhaps one of the most important aspects of macromolecular dynamics accessible at the LWTS is the natural connection between such picosecond and nanosecond motions on the Å length scale in native proteins with molecular dynamics (MD) simulation studies. In order for MD to realize its full capability as a predictive tool, accurate interatomic potentials must be used, and neutron scattering data are ideally suited verifying such potentials because of the ease with which neutron scattering spectra can be calculated and compared with experiment. This approach will be even more powerful with future increases in computing power that will make it routinely possible to 'tune' potentials by comparison with neutron scattering results. This combined use of MD and neutron scattering spectroscopy has already made a significant impact in the area of relatively complex disordered systems. Examples include

biologically important interacting ligands in solution, as well organic and inorganic materials that are increasingly important to modern technologies, allowing a revolution in materials design and optimization.
The large increase in flux available at the LWTS will make it possible to work in areas of molecular biology where a systematic study of a large variety of similar molecules under slightly different conditions is required. For much of biology, it is the small differences between closely related systems, which are of importance. The large increase in flux on the LWTS will make such systematic studies possible for the first time, enabling molecular biology and biotechnology to take a much fuller advantage of the power of the neutron scattering to provide detailed microscopic views of both structure and dynamics.
Also the enormous increase in available neutron flux provided by the SNS along with the widening of the dynamic range accessible by its spectrometers, particularly those at the LWTS, will have a substantial impact on several areas of chemistry, such as catalysis, inorganic and materials chemistry. Slow molecular diffusion of some relatively large molecules in a wide variety of catalytic materials will be accessible on time scales complementary to those of PFG NMR methods, as will the motion of charge carrying ions in energy-storage materials. A most prominent example of the latter is the diffusion of Li ions in Li battery materials. Proton transfer dynamics is of great importance in a wide variety of chemical and biological processes, and has only recently been observed by neutron spectroscopic methods in such systems as proton conductors and transition metal polyhydrides. Both the increase in flux and dynamic range should make it possible to envisage the observation of proton transfer in catalytic reactions such as hydrogenation by quasi-elastic neutron scattering. This would provide unprecedented details into the mechanism of such reactions. Given the fact that some spectra can be obtained in minutes at the SNS spectrometers it will then be possible to study slow dynamic processes as a function of time, e.g. by varying the sample conditions. For example, time-dependent structural studies are currently being carried out on topics such as zeolite crystallization. It is conceivable that this type of work can be extended to include motions of the structure directing agents used in this process.

**Workshop on Chemical Spectroscopy, Protein Folding Dynamics and Polymer Dynamics**

**The 200 neV back-scattering instrument proposed for the Long Wavelength Target Station: Systematic studies over a large dynamic range**

Tuesday October 10 and Wednesday October11 2000
8:30 AM to 17:00 PM
Argonne National Laboratory, Advanced Photon Source, Conference Room E1100

Tuesday October 10

| | |
|---|---|
| 08:00 – 08:30 | Welcome Working Breakfast |
| 08:30 – 08:40 | H.N. Bordallo and J. Kent Blasie – Plans for the day |
| 08:40 – 09:00 | J. Richardson (ANL-IPNS) – Overview<br>"LWTS: Overview, Instrumentation and the Science case" |
| 09:00 – 09:30 | J. Carpenter (ANL-IPNS)<br>"LWTS: Concept Development" |
| 09:30 – 10:00 | K.W. Herwig (SNS- ORNL)<br>"The Backscattering Spectrometer on the SNS High Power Target Station" |
| 10:00 – 10:15 | Coffee Break |
| 10:15 – 11:00 | H.N. Bordallo (ANL-IPNS)<br>"Monte-Carlo simulations of a high-resolution inverse geometry spectrometer on the SNS - Long Wavelength Target Station" |
| 11:00-11:45 | R.E. Lechner (Hahn-Meitner-Institut/BENSC)<br>"Ionic conductors and biomolecules : Functionally relevant motions from QENS" |
| 11:45 – 13:00 | Working Lunch |
| 13:00 – 13:45 | J. Kent Blasie (University of Pennsylvania - Dept. of Chemistry)<br>"Key Problems in Membrane Protein Intramolecular Dynamics" |
| 13:45 – 14:30 | Jyotsna Lal (ANL-IPNS)<br>"Examples of Soft Systems probed by the technique of Neutron Spin Echo" |
| 14:30-15:15 | D. J. Tobias (University of California, Irvine- Dept. of Chemistry)<br>"Neutron spectroscopy and molecular dynamics of proteins and membranes" |
| 15:15-15:30 | Coffee Break |
| 15:30-16:15 | D. G. J. Kearley (Delft University of Technology - IRI)<br>"Rotational tunnelling: What have we learnt? What will we learn? What use is it?" |
| 16:15-17:00 | Discussion on potential scientific achievements using inelastic and quasi-elastic neutron scattering |
| 19:00 | Workshop dinner at the Guest House |

Wednesday October 11

08:00 – 08:25 Working Breakfast

08:25 – 08:30 H.N. Bordallo and J. Kent Blasie – Plans for the day

08:30 – 09:15 Z. Bu (NIST)
"Dynamics changes in the molten globule-native folding step studied by quasielastic neutron scattering"

09:15 – 10:00 K. Gawrisch (NIH-NIAAA, LMBB)
"Structure and dynamics of polyunsaturated neural receptor membranes"

10:00 – 10:45 M. L. Klein (University of Pennsylvania – Dept. of Chemistry)
"Computer Simulation of Membranes and Membrane Proteins"

10:45 – 11:00 Coffee Break

11:00 – 11:45 J. Eckert (LANSCE-LANL)
"High Resolution Spectroscopies in Chemistry"

11:45 – 12:30 G.F. Strouse (University of California, SB- Dept. of Chemistry)
"Correlating structure and dynamics in materials using spectroscopic probes"

12:30 – 13:30 Working Lunch

13:30 – 14:15 B. Frick (ILL)
"Neutron backscattering spectroscopy for soft matter - recent results and perspectives"

14:15 – 17:00 Groups breakout:
How potential scientific achievements using inelastic/quasi-elastic neutron scattering could be enabled by a Long Wavelength Target Station at the SNS?

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Workshop on Macromolecular Single Crystal Diffraction at
The Spallation Neutron Source

## Executive Summary

On December 18th and 19th of 2000 a workshop, co-sponsored by the Spallation Neutron Source (SNS) project and the National Aeronautics and Space Administration (NASA), was held in Knoxville to discuss the future use of the SNS for macromolecular single crystal neutron diffraction. This workshop brought together for the first time in the United States representatives of both the biological neutron diffraction and microgravity crystal growth communities. At the conclusion of the workshop, the following recommendations were made: 1) single crystal biological instrumentation should form an integral part of the SNS instrument suite; 2) all funding options should be pursued and supported to facilitate the development of two instruments for macromolecular crystallographic neutron diffraction studies at SNS; and 3) the workshop participants support efforts to fund the Long Wavelength Target Station (LWTS) and the proposed complex biological system diffraction station; in addition on behalf of the group, the workshop organizers should submit a letter of intent to the Experimental Facilities Advisory Committee (EFAC), proposing a high-resolution macromolecular diffraction instrument at the High Power Target Station (HPTS).

## Introduction

Neutron diffraction provides important and unique information for macromolecular structure–function studies. Hydrogens comprise roughly half the atoms of biological materials such as proteins and DNA, and hydrogen ions supply the primary motive force in the molecular actions of such fundamental biological processes as metabolism and reproduction. The ability of neutrons to reveal the positions of hydrogens even at moderate resolution (2.5 Å) is the foundation of the scientific justification for neutron diffraction of biological samples. Although other techniques may provide complementary information, none do it at such moderate resolution levels nor as conclusively as neutrons. In order to reassert the value of neutron crystallography and the justification for macromolecular neutron diffraction experimentation and to explore the role of the SNS in future neutron diffraction studies a two-day workshop was organized.

In addition to presentations on the scientific role and justification for biological neutron crystallography, other presentations on the first day included an overview of the Spallation Neutron Source project, scientific advances with macromolecular neutron diffraction, recent developments in neutron detectors and beam lines world wide, and the role of microgravity in the growth of crystals suitable for neutron diffraction experiments.

The potential role of macromolecular neutron diffraction at SNS has not been recognized, as illustrated even in local news media reports. Therefore, an important mission of the workshop was to inform both crystallographers and other biologists interested in structure-function studies of biomolecules on the merits of neutron diffraction studies, as a preamble to more widespread dissemination of information. There is a gathering confluence of events that will provide opportunities to grow large crystals on the International Space Station (ISS), and to collect data at high flux neutron sources. These include the Institut Laue Langevin (ILL), the Rutherford Appleton Laboratory's spallation source (ISIS), the SNS, and the proposed European Spallation Source (ESS). This potentially exciting new era in biological neutron diffraction studies is capturing the attention of governments and funding organizations worldwide.

## Recommendations

Three primary conclusions were reached from the workshop presentations and discussions:

1.) The landscape of macromolecular neutron diffraction has undergone a significant positive alteration in the past three years. The consequence of this alteration is a significant shift in the conditions that existed at the time of the Biological and Environmental Research Advisory Committee (BERAC) Report of 1998. That committee suggested a de-emphasis of neutron diffraction in part because of the probability of more readily obtaining ultra-high resolution structural information from X-ray data collected at 3$^{rd}$ generation synchrotron sources; the advent of GHz and higher frequency NMR devices capable of generating interpretable spectra from larger proteins; the absence of large crystals suitable for neutron diffraction and the lack of reliable means to grow such crystals; and the less than optimal combination of neutron source flux and detector sensitivity that results in multi-week data collection times even from proteins with small unit cells. However, it is the consensus of this workshop that the 1998 report recommendations for neutron diffraction studies have been superseded by new scientific findings and instrumentation developments.

a.) In contrast to the views cited in the BERAC report, the potential for using neutron sources to determine positions of hydrogens has become increasingly attractive. A survey of X-ray structures in the Protein Data Bank suggests that crystals of sufficient quality to diffract to > 0.9 Å resolution occur only in ~1% of proteins sampled [Langan, Myles, Timmins]. A further complication of high resolution 3$^{rd}$ generation synchrotron data collection is radiation damage to crystals, necessitating attenuation of high brilliance sources, thus reducing the ability to collect ultra-high resolution X-ray diffraction data needed to establish positional information on the hydrogen atoms in these molecules. In crystals that do diffract to ultra-high resolution, significantly more information on solvent structure and hydrogen positions can be derived from moderate resolution neutron data (2.3-2.5 Å) than from X-ray diffraction data (0.9 Å) [Myles and Langan]. A more conservative view suggests that static hydrogen positions will come in increasing numbers from ultra-high resolution X-ray data, but that X-ray data will never match neutrons in the positioning of partially mobile hydrogens [Helliwell]. Support for both positions is seen in crystals of concanavalin A where deuterium atom positions and orientations can be seen in a greater number of water molecules with neutrons (60 $D_2O$'s) than with synchrotron X-rays (10 $H_2O$) [Habash *et al.,* 1999]. Thus, the neutron approach is more efficient even with the data collection occurring at room temperature [Helliwell. Positions of non-exchanged hydrogens can be determined in crystals that have been soaked in $D_2O$ to replace labile hydrogens and solvent. The positions of these hydrogen atoms can be established from moderate resolution data (2.3 Å) by examining maps of negative density (corresponding to the negative scattering length of hydrogen) [Niimura *et al*., 1997]. The positions of deuterium atoms appear as positive density in these same maps. Neutron diffraction also presents the possibility of determining meaningful thermal parameters for H and D atoms, for high-resolution structures. For example, it may be possible to correlate these thermal parameters with hydrogen-bonding strengths. [Koetzle]

b.) As a result of research supported by both NASA and the European Space Agency (ESA), the production of crystals sufficiently large for neutron diffraction studies is considered an attainable goal. Growth of crystals with 2 mm x 1.5 mm x 1 mm (or larger) volume is now common for an increasing number of proteins [Carter]. With the commissioning of the International Space Station a permanent venue now exists that should be available to crystallographers interested in using microgravity to enhance crystal growth [Carter and Snell]. Based on previous microgravity crystal growth experiments, and the availability of a controlled environment for extended duration missions, it is estimated that ~90% of proteins crystallized on orbit will have the potential to reach the 1 mm x 1mm x 1mm size range needed for experiments on current and future neutron sources.

c.) Significant improvements have been realized in neutron detectors. Increased sensitivity of detectors has resulted in decreased data collection times. Detector and beamline developments in Japan, at Grenoble (ILL), and at Los Alamos (LANSCE), have resulted in full data sets being collected within 10 days, even 24 hrs, rather than months [Niimura, Myles, Langan].

d.) Thus, although neutron data collection will never be described as high-throughput, based on flux projections and detector developments, it should be possible at SNS to collect a complete neutron diffraction data set from a crystal of 0.5 mm x 0.5 mm x 0.5 mm with a longest unit cell dimension of 100 Å within 1 week of beam time. This is comparable to the size of crystal and time period needed to complete data collection with typical in-house X-ray systems.

Because of these advances and predictions, and in the absence of dedicated equipment planned for macromolecular crystallography, it was the consensus of the participants that single crystal biological instrumentation must form an integral part of SNS instrument station planning.

2.) Future planning for the SNS should include two different protein crystal diffraction instruments. The first instrument, capable of collecting high-resolution data from crystals with a maximum unit cell length of 100 Å, should be built at the High Power Target Station (HPTS). A second instrument capable of collecting medium-high resolution data from crystals with unit cell axes of up to 250 Å should be built at the Long Wavelength Target Station (LWTS). This instrument should have additional capabilities for membrane, fiber, and low-resolution diffraction studies.

a.) These two instruments will address different crystallographic research. The HPTS instrument will provide essential high-resolution (1-1.5 Å) proton (deuteron) and water structure data for moderate sized proteins that cannot be determined using other diffraction or resonance techniques. This device will provide a means for determining mechanisms and biochemistry of novel enzymes and other proteins discovered in functional/structural genomic studies [Helliwell, McRee, Stallings]. The LWTS instrument will provide a venue to answer fundamental questions about large proteins and complex assemblages at medium- high resolution. The LWTS instrument will be able to determine the positions of the protons/deuterons in complex biomolecules like the nucleosome core particle, the fundamental building block of the chromosome, but may fall short of being able to pinpoint the position of the catalytic proton related to protein synthesis in the ribosome (unit cell dimensions exceeding 300 Å).

b.) Results from questionnaires distributed by the macromolecular beamline group at the spallation source at Los Alamos suggest that when fully commissioned the instrument there will be capable of meeting the needs of about 20% of the potential users in the US [Langan & Schoenborn, 1999]. The questionnaire results are based on present interest and do not reflect potential increased usage as a new generation becomes aware of the value of neutron diffraction data. Workshop participants suggested that similar levels of interest in synchrotron data collection existed prior to widespread recognition of their utility [Helliwell, Minor].

c.) Shared instrumentation with the small molecule crystallographic community at HPTS is not a viable option. The geometry of this device will not allow the resolution of data from large unit cell crystals. In addition, ancillary equipment at the small molecule instrument will restrict the sample and detector geometry required to collect macromolecular data [Zhao, Schultz]. These devices include furnaces and refrigerators for high and low temperature studies, magnets, and high-pressure chambers.

Based on the proposed mission and potential demand for instrumentation (and concomitant absence of similar instrumentation elsewhere) it was the sense of the workshop participants that all funding options be pursued and supported to facilitate the development of two instruments for macromolecular crystallographic neutron diffraction studies at SNS.

3.) Current proposals submitted to NSF for funding a LWTS at SNS include the second macromolecular device described earlier [Mason]. All efforts should continue to ensure that the LWTS is funded and constructed. For the proposed HPTS instrument, it is possible that an Instrument Development Team (IDT) will be needed to ensure its inclusion at SNS. The workshop participants charged the organizers with the task of submitting a letter of intent (LOI) for the HPTS instrument to the SNS Experimental Facilities Advisory Committee (EFAC), and the identification of potential funding agencies. The next opportunity to submit the LOI is March 2001.

In addition to these conclusions, it was felt that educational and outreach activities should to be undertaken to acquaint a new generation of crystallographers with the scientific merits of neutron diffraction and with the advances that have been made in recent years. To this end, the workshop participants will seek to make presentations at various national societies, including the American Crystallographic Association and the Protein Society. Plans for the ACA 2001 annual meeting will include a special interest group session on macromolecular neutron crystallography.

## Summary of Workshop Presentations

### *SNS and Advances in Neutron Crystallography*

The SNS will have a 2 megawatt power output: this is about half the integrated neutron flux of ILL, but 50 times greater peak flux and significantly greater (12X) than that of the current highest-flux spallation source, ISIS, in England. The design of the SNS should permit power upgrades without significant future expenditures. Site excavation has been completed, foundations are being poured, and the first neutrons are expected in June of 2006. One target station, the high power target station (HPTS) is funded integral to the project, and funding is being sought from NSF for the construction of a second target station, the Long Wavelength Target Station (LWTS). Further information is available at the SNS website (www.sns.gov).

At present, no instrumentation for macromolecular crystallography is in development. The process for externally funded instrument selection at SNS starts with the formation of an instrument development team (IDT), the submission of a letter of intent from the IDT, followed by a decision from the Experimental Facilities Advisory Committee (EFAC) on recommending a conceptual design study of the proposed instrument. Alternatively, acquisition of a macromolecular instrument at the HPTS of SNS could be included in the competition for the initial suite of instruments. Instrumentation for a macromolecular crystallography beam line is currently included in proposals for funding the LWTS.

The scientific case for neutron diffraction was presented by John Helliwell whose research into the basis of sugar recognition by concanavalin A has encompassed extensive synchrotron X-ray and neutron data collection. Europeans view the SNS as becoming the state of the art facility for neutron users, and as a justification for the construction of the European Spallation Source (ESS). As Europe watches developments with SNS, continued upgrades of existing neutron facilities are underway both in Britain and on the continent. These upgrades include a new area detector for the D19 beam line at ILL, and upgrades of neutron Laue time-of-flight instruments at ISIS, indicating a movement toward more protein crystallography for detailed analysis of hydrogen bonding and catalytic site structure.

The traditional basis for neutron diffraction is the ease with which accurate positions of hydrogens/deuterons can be determined in crystals of macromolecules. Both deuterium and oxygen scatter similarly in neutron diffraction experiments. Thus, solvent position and proton exchange can be readily identified from neutron diffraction data. A more recent rationale for neutron diffraction can be seen in the comparative diffraction data for concanavalin A between an ultra-high resolution X-ray cryostructure and a medium resolution neutron room temperature structure. The neutron data were collected at ILL from a $D_2O$ soaked crystal of 3 mm x 2 mm x 1 mm. Data were collected in 10 days and were 89% complete to 2.4 Å. The neutron structure provided six times the number of well-determined waters (position and orientation) compared with the ultra-high resolution X-ray data (0.9 Å). Thus, neutron diffraction determines bound waters more efficiently, and it also should provide the primary means of identification of the positions of somewhat mobile waters in a protein structure [Habash *et al.,* 1999].

Neutron diffraction data should have a pre-eminent role in assisting future modeling studies and computational biology, by providing a structural data base for understanding the role of solvent in ligand interaction, and providing further needed information to understand the thermodynamics of ligand recognition from structural data. Such studies will have a direct impact on rational drug design, providing more accurate and complete molecular structures. Thermal parameter values for H and D atoms may also be assigned based on neutron diffraction data, for proteins that diffract to high resolution. To fully understand the water structure and proton exchange rates of native molecules low temperature X-ray data must be compared with room temperature neutron data. Even at ultra-high resolution, X-rays are not good at distinguishing water molecules from monovalent cations (sodium, ammonium, potassium). It will not be possible to understand things like RNA folding and catalysis, or DNA bending unless cation positions are identifiable.

Peter Timmins reported that unique information on the location of H atoms and water has been obtained by neutron fiber diffraction of biological polymers, including cellulose, hyaluronic acid, filamentous viruses, and DNA. For example, individual water positions along the DNA strand have been refined even at the low resolution of 3 Å. Other significant diffraction information can be obtained at low resolution including the localization of surfactants added to proteins. The detergent structure in integral membrane proteins has not been possible to determine in X-ray diffraction studies owing to the disorder of the surfactants in the unit cell. It has now been possible to resolve the detergent structure in crystals of OmpF porin of *E. coli* using neutron contrast matching studies [Pebay-Peyroula *et al.*, 1995]. Given that 40% of the genome is membrane-bound proteins, which are extremely difficult to crystallize, the strategic importance of such neutron studies in revealing the interactions of proteins and detergents cannot be emphasized enough.

Improvements in neutron sources, detector design and interpretation of multi-wavelength diffraction have improved the speed with which data can be collected. Detector improvements include the neutron image plate, which is currently in use in both Japan and Europe, a neutron area detector with 1mm pixel size being developed at Oak Ridge, and detector research at Brookhaven National Laboratory for the SNS project. Nobuo Niimura presented rubredoxin data (unit cell axes 34 Å x 35 Å x 44 Å; crystal size 2.6 mm x 1.7 mm x 1.0 mm) collected with a neutron image plate in 11 hrs. The resolution of the data is 1.5 Å and the refinement of the hydrogen positions at 1.5 Å is currently underway. At the Japanese Atomic Energy Research Institute (JAERI), the rule of thumb for data collection from macromolecular crystals is that each axis must be less than 100 Å. However, longer unit cell lengths can be accommodated if the other axes are smaller (i.e., 50 Å x 50 Å x 200 Å). The volume of the crystal must be ≥ 2 $mm^3$, so 1.5 mm x 1.5 mm x 1.0 mm is around the current minimum acceptable crystal volume. Construction has started on the new BIX-4, the performance of which will be at least 3 times better than the present BIX-3. The necessary crystal volume should be ~1/3 that of BIX-3.

In addition to steady state neutron sources, several spallation sources are planning to contribute to biological neutron diffraction research. ISIS, at the Rutherford Appleton Laboratory, does not have a protein crystallography instrument in place, but the facility is currently undergoing upgrades consisting of a second target station, which will include such an instrument. In this country, the Protein Crystallography Station (PCS) at the Lujan Center source at the Los Alamos Neutron Sciences Center (LANSCE) produced its first neutron beam on a sample in the week prior to the conference. The PCS is a new diffraction instrument for protein crystallography, fiber, and membrane diffraction. This instrument, with a partially coupled water moderator, uses a large cylindrical position sensitive detector for collecting data. The detector has an active area of 3000 $cm^2$ and a resolution of 1.3 mm with a counting rate over 1 million counts/sec. A call for proposals at the LANSCE PCS will be issued in 2001. An advisory team of structural biologists is in place for the LANSCE PCS, and contacts with this group will be pursued.

As a result of microgravity studies supported by both NASA and the ESA, production of crystals sufficiently large for neutron diffraction studies could become commonplace. Dan Carter reported reliable growth of crystals with the 1-$mm^3$ volume needed for current neutron diffraction experiments for even such problematic proteins as bacteriorhodopsin. With the commissioning of the International Space Station a permanent venue now exists that should be available to crystallographers interested in using microgravity for crystal growth. It is estimated that with the extended duration missions on the ISS, ~90% of proteins crystallized on orbit will reach the 1-$mm^3$ size range needed for neutron diffraction experiments. For example, on flight STS-89, a crystal of ferritin was grown with a volume of 10 $mm^3$. Neutron diffraction data to 2.7 Å has been collected at ILL from this crystal, 1 Å higher resolution than the diffraction limit of ground grown crystals. In studies reported by Eddie Snell, microgravity grown crystals are consistently larger and more physically perfect than those grown terrestrially. This physical perfection takes place on both short and long-range scales, and has been quantified using X-ray diffraction rocking-curve studies. In contrast, despite early optimistic claims cited in the BERAC report, the evidence suggests that crystals of sufficient quality to diffract to > 0.9 Å for determination of hydrogen positions occur only in ~1% of proteins sampled.

One significant application for proteins structures determined by neutron diffraction will come in the area of rational drug design. Chris Dealwis pointed out the practicality of this approach, discussing the problems associated with finding an agonist of angiotensin to bind with rennin, and how this study carried over to drug discovery of anti-HIV proteases. Both rennin and HIV protease belong to the general class of aspartic proteases, so named for the aspartic acid moieties, which are an integral part of the catalytic site. A solvent molecule bound tightly to both aspartate carboxyl groups is presumed to take part in the catalytic mechanism. Currently proposed mechanisms are largely based on X-ray inhibitor structures, but the assignment of protonation states to the catalytic groups during the reaction differ. Since the active-site H atoms cannot be located by current X-ray analyses, their putative positions have so far been inferred from the local geometry of surrounding polar atoms. Thus, locating the crucial protons at the active site will provide important information to firmly establish the catalytic mechanism.

Enzyme structure and mechanism continue to be fertile ground for neutron crystallography, from the seminal study of the catalytic triad of trypsin [Kosiakoff & Spencer, 1981], to new studies of aspartic proteases. A recent report of the neutron diffraction structure of the fungal aspartic protease endothiapepsin by Cooper and Myles (2000) is a research milestone for several reasons. It represents the largest protein solved by neutron diffraction methods (33 kDa), and, by establishing the positions of the catalytic protons, represents a route to the development of more effective inhibitors to aspartic proteases. The endothiapepsin structure is the beginning of what may be one of the more significant roles played by neutron diffraction studies.

***Discussion of Neutron Instrumentation at SNS***

Because important information will be provided by studies of both smaller and larger proteins the development of two instruments were proposed, one that would be capable of resolving atomic positions for protein crystals with maximum unit cell axes of ~100 Å, and another device to resolve medium-high resolution data from crystals with maximum unit cell axes of ~250 Å. Projected flux and wavelength models for SNS suggest that an instrument at HPTS coupled to a chopper to utilize neutron wavelengths of 0.9-2.1 Å will meet the parameters of the first device. When fully commissioned, this instrument should be capable of collecting a complete data set of atomic resolution data from a crystal 0.5 mm x 0.5 mm x 0.5 mm in less than a week. This translates into ~30-50 new neutron structures a year, or nearly fivefold greater than the neutron structures cataloged in the Protein Data Bank. The HPTS device could augment the role of a variety of high throughput programs, producing more readily interpretable molecular mechanism data for new enzymes discovered by Structural Genomics.

The instrument for larger unit cells should provide diffraction data to elucidate atomic position models of large proteins, protein-protein complexes and protein-nucleic acid assemblages. Plans for this instrument are included in the proposal for the LWTS submitted to NSF (www.sns.anl.gov/LWTS/NSF_LWTS_Proposal.pdf). The inclusion of both instruments at SNS represents the minimum fulfillment of potential needs of the macromolecular diffraction community. Further instruments may be needed if the use of SNS for neutron diffraction studies follows a similar trajectory as the use of X-rays at synchrotrons.

The second day was dedicated to detailed discussions of the needs to be met for successful macromolecular crystallography at SNS. Discussions of detector and sample position geometry made it clear that a dual use (small-molecule and macro-molecule) crystallography instrument will not fit the needs of either community. The needs of the small-molecule community include furnaces and refrigerators, as well as magnets and high-pressure devices that will interfere with the positioning of neutron detectors for macromolecular data collection. Because of the large differences in reciprocal lattice spacing of small molecules and macromolecules, the optimization and placement of detectors for small and macromolecular data collection are quite different. Continuing research and development in both neutron moderators and detectors were encouraged by the group. The workshop participants noted that the budget for neutron detectors is under great stress at SNS. Developments in moderators and detectors over the next several years will provide the most beneficial and cost effective approaches for realizing additional improvements in data collection times, especially for the instrument at the LWTS.

## Recommendation Summary

1) Single crystal biological instrumentation should form an integral part of the SNS instrument suite.

2) All funding options should be pursued and supported to facilitate the development of two distinct instruments for macromolecular crystallographic neutron diffraction studies at SNS.

3) The workshop participants support efforts to fund the Long Wavelength Target Station (LWTS) and the proposed complex biological system diffraction station.

4) The workshop organizers should submit a letter of intent to the Experimental Facilities Advisory Committee (EFAC), proposing a high-resolution macromolecular diffraction instrument at the High Power Target Station (HPTS). Planning should begin now to identify agencies for funding the development of this instrument.

5) Educational and outreach activities should be undertaken to acquaint a new generation of crystallographers of the scientific merits of neutron diffraction.

6) Develop plans for the ACA 2001 annual meeting to include either a workshop or a special interest group session on macromolecular neutron crystallography, if feasible.

7) Inform LANSCE PCS advisory board members of the SNS Macromolecular Crystallography Workshop and invite their participation.

**Submitted on behalf of the workshop by:**

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Chris Dealwis, The University of Tennessee, Department of Biochemistry and Cellular Molecular Biology

B. Leif Hanson, The University of Tennessee/ORNL Graduate School in Genome Science and Technology

John R. Helliwell, The University of Manchester, Department of Chemistry

Jinkui Zhao, Oak Ridge National Laboratory, Spallation Neutron Source Project

**Appendix**
**SNS Macromolecular Crystallography Workshop Participants**

**SNS/LWTS Neutron Protein Crystallography**
**December 18-19, 2000 UT Conference Center**

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| Uchiki, Tomoaki | BCMB UTK-grad student | UTK, BCMB |
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**Interested, Could Not Attend**

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**Agenda**

**S N S Macromolecular Crystallography Workshop**

Monday, December 18

**Morning Session Chair: Chris Dealwis**

| | |
|---|---|
| 8:00-9:00 a.m. | Registration and Continental Breakfast |
| 9:00-9:10 | Welcome / Opening Remarks (Gerry Bunick) |
| 9:10-9:40 | Overview of SNS Project (Thom Mason) |
| 9:40-10:30 | Neutron Macromolecular Diffraction (John Helliwell) |
| 10:30-11:10 | Biological Applications of Neutron Diffraction at the ILL<br>High Resolution Diffraction (Dean Myles)<br>Fiber and Low-Resolution Diffraction (Peter Timmins) |
| 11:10-11:30 | **Coffee Break** |
| 11:30-12:30 | Neutron Diffraction in Japan/ Neutron Area Detectors<br>(Nobuo Niimura and Ichiro Tanaka) |
| 12:30-1:00 | Neutron Instrumentation and Biological Research at ISIS<br>(Charles Wilson) |
| 1:00-2:00 | **Lunch Break** |

**Afternoon Session Chair: Gerry Bunick**

| | |
|---|---|
| 2:00-2:40 | The NeutronProtein Crystallography Station at LANSCE<br>(Paul Langan) |
| 2:40-3:10 | SNS Detector Development at ORNL (Donald Hutchinson) |
| 3:10-3:40 | Microgravity Crystal Growth and Commercial Neutron Macromolecular Crystallography (Dan Carter) |
| 3:40-4:00 | **Coffee Break** |
| 4:00-4:30 | Exploiting Neutrons for Drug Design (Chris Dealwis) |
| 4:30-5:00 | Microgravity and Neutron Crystallography (Eddie Snell) |
| 5:00-5:30 | Options for Macromolecular Diffraction Beamlines at the SNS<br>(Jinkui Zhao) |

**Tuesday, December 19**

**Session Chair: Jinkui Zhao**

| | |
|---|---|
| 8:00-9:00 | Continental Breakfast |
| 9:00-11:00 | Discussion: Instrument Selection and Design (Organizing Committee and Participants) |
| 11:00-11:20 | **Coffee Break** |
| 11:20-1:00 | Discussion: Instrument Selection and Design (Organizing Committee and Participants) |
| 1:00-2:00 | **Lunch** |
| 2:00-3:30 | Discussion: Instrument Administration and Funding (Organizing Committee and Participants) |

## References and Recent Publications


Bon C., Lehmann M.S. and Wilkinson C. (1999) Quasi-Laue neutron-diffraction study of the water arrangement in crystals of triclinic hen egg-white lysozyme. *Acta Cryst.* D**55**, 978-987.

Cooper J.B. & Myles D.A. (2000) A preliminary neutron Laue diffraction study of the aspartic proteinase endothiapepsin. *Acta Cryst*. D**56**:246-248.

Habash J., Raftery J., Nuttall R., Price H.J., Wilkinson C., Kalb (Gilboa), A.J. and Helliwell J.R. (2000) Direct determination of the positions of the deuterium atoms of the bound water in concanavalin A by neutron Laue crystallography.
*Acta Cryst*. D**56**, 541-550.

Kossiakoff, A.A. & Spencer, S.A. (1981) Direct determination of the protonation states of aspartic acid-102 and histidine-57 in the tetrahedral intermediate of the serine proteases: Neutron structure of trypsin. *Biochemistry* **20**: 6462-6474.

Langan P. & Schoenborn B.P. (1999) Need for neutron diffraction instruments. *Science* **286**:1089

Langan P., Lehmann M., Wilkinson C., Jogl G. and Kratky C. (1999) Neutron Laue diffraction studies of coenzyme cob(**II**)alamin. *Acta Cryst.* D**55**, 51-59.

Myles D.A., Bon C., Langan P., Cipriani F., Castagna J.C., Lehmann M.N. and Wilkinson C. (1998) Neutron Laue diffrac
tion in macromolecular crystallography. *Physica B* **241-243**:1122-1130.

Niimura N., Minezaki Y., Nonaka T., Castagna J.C., Cipriani F., Hoghoj P., Lehmann M.S., Wilkinson C. (1997) Neutron Laue diffractometry with an imaging plate provides an effective data collection regime for neutron protein crystallography. *Nat Struct Biol*., **4**:909-914.

Niimura N. (1999) Neutrons expand the field of structural biology. *Current Opinion in Structural Biology* **9**:602–608

Pebay-Peyroula E., Garavito, R.M., Rosenbusch, J.P., Zulauf, M., and Timmins P.A. (1995) Detergent structure in tetragonal crystals of OmpF porin. *Structure* **3**: 1051-105

Shu F., Ramakrishnan V., and Schoenborn B.P. (2000) Enhanced visibility of hydrogen atoms by neutron crystallography on fully deuterated myoglobin. *Proc Natl Acad Sci U S A* **97**:3872-3877

Neutron Vibrational Spectrometer Planning Workshop – John Larese
February 1-2, 2002 – Knoxville, TN

The Basic Energy Sciences Advisory Committee (BESAC) Subpanel Report on Neutron Scattering February 2000 chaired by M. Blume in early 2000 (charged with looking at the crisis faced by the neutron scattering community upon closure of the HFBR at BNL) included a recognition that future growth area in the neutron scattering community was likely to be in the area broadly defined as Physical Chemistry (including Materials Chemistry, Chemical Physics, Inorganic chemistry, Solid State Chemistry and activities related to nanoscience). During the subpanel meetings it was established that vibrational spectroscopy (principally Infrared and Raman spectroscopy) is an extremely valuable tool for a large segment of the scientific community. Furthermore, it was pointed out that this community represented nearly 50% of the neutron scattering effort in Europe. The instrument TOSCA at the Rutherford Appleton Spallation Neutron facility ISIS was used to demonstrate the power of spallation neutron sources in the study of vibrational spectroscopy. J. Z. Larese was a member of that BESAC panel.

A presentation by B. S. Hudson Syracuse University and J. Z. Larese (University of Tennessee/ Oak Ridge National laboratory) was made to the Spallation Neutron Source (SNS) Experimental Facilities Advisory Committee (EFAC) in October 2001 suggesting that a next generation instrument for performing simultaneous vibrational spectroscopy and diffraction be considered as an instrument for construction at SNS. Encouraged by strong words of support by the EFAC committee, a working group was contacted and a workshop organized by J. Z. Larese and B. S. Hudson and held in Knoxville February 2002 to explore the merits of building such a spectrometer. The following individuals attended the workshop:

- T. Arnold, Post Doctoral Fellow, ORNL
- C. Brown, Staff Member, NIST Neutron Group
- R. Cook, Graduate Student, UTK Chemistry
- L. Daemen, Staff Member, LANSCE
- J. Eckert, Staff Member, LANSCE
- B. Hudson, Professor Chemistry, Syracuse
- G. Kearley, Professor Technische, Univ. Delft
- J. Larese, Joint Faculty Professor UTK/ORNL Chemistry
- S. Parker, Staff Member ISIS RAL
- J. Tomkinson, Head Spectroscopy Group ISIS, RAL
- J. Turner, Asst. Professor, Chemistry UTK
- M.A. White, Professor Chemistry Physics Dept. Dalhousie Univ.
- M. Zoppi, Staff Member CNR, Italy

The workshop ran for one and a half full days (Letter of Invitation and Agenda are below) and resulted in an unanimous vote to form an Instrument Design Team called VISION (with all the participants as charter members) and that a letter of Intent (LOI) be sent to SNS to express that desire (LOI in separate document). As a result a principal investigator (J. Z. Larese) was identified and a presentation was made to SNS EFAC in April 2002 requesting approval for the formation of an Instrument Development Team (IDT). EFAC's recommendation was that the IDT should be formed and that the development of a full proposal for the vibrational spectrometer be prepared.

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January 18, 2002

Dear Colleagues:

Thank you again for agreeing to participate in the workshop designed to write a letter of intent to SNS to develop an instrument focused on Chemical Spectroscopy. If you haven't arranged your travel plans as of I hope you'll do that soon. The meeting will be held at the Radisson in Downtown Knoxville. The activities will begin on Friday, February 1 morning at 9:00 and be completed by 12:30 on Saturday, February 2. There is the possibility that we could arrange a tour of the SNS site on Saturday afternoon for those of you who are interested.

Maria Fawver will be handling the important tasks associated with this meeting. Please communicate your travel plans with her as soon as you have made them. Expect to take a taxi/limo from the McGhee-Tyson Airport to the Radisson. She can be reached via e-mail at (mfawver@utk.edu) or phone or fax but, don't hesitate to contact me (jzl@utk.edu) or Bruce Hudson (bshudson@syr.edu) if you have any problems or concerns that we need to attend to. Please come prepared to give a 15-minute description of the type of science you're interested in, especially as it applies to this area. Also let us know if we need to provide equipment for more than overhead or power point presentations. Please let Ms. Fawver know.

Once again I look forward to this inaugural meeting as the beginning of a successful adventure in neutron science.

Sincerely yours,

John

p.s. Please express your interest to visit the SNS site to Maria since we will have to make prior arrangements with ORNL/SNS at least a week in advance

**<u>SNS Chemical Spectrometer Workshop Agenda</u>**

**February 1, 2002** **Radisson Hotel, Knoxville TN**

| | |
|---|---|
| **8:00-9:00** | **Continental Breakfast** |
| **9:00-9:05** | Welcome (J. Roberto, Assoc. Laboratory Director for BES, ORNL) |
| **9:05-9:10** | Statement of Objective (Larese) |
| **9:10-10:40** | Participants 15 minute Science Interest Statement |

9:10 C. Brown (NIST)
9:25 L. Daemen (LANSCE, LANL)
9:40 B. Hudson (Syracuse Univ.)
9:55 M. Johnson (Institute Laue-Langevin, Grenoble)
10:10 G. Kearley (Delft)
10:25 J. Z. Larese (UTK/ORNL)

| | |
|---|---|
| **10:40-11:00** | Break |
| **11:00-12:00** | Participants 15 minute Science Interest Statement Con't |

11:00 S. Parker (RAL/ISIS)
11:15 J. Tomkinson (RAL/ ISIS)
11:30 J. F. C. Turner (UTK)
11:45 M. A. White (Dalhousie Univ. Canada)
12:00 M. Zoppi (CNR – Italy)

**12:15-3:00** Lunch

**13:30-14:30** Review Capabilities / Short comings Current Instruments

| | | |
|---|---|---|
| ISIS | TOSCA | Tomkinson/Parker |
| LANSCE | FDS | Daemen |
| NIST | FANS | Brown |

**14:30-15:30** Review Modeling / Data Reduction Software Capabilities
Kearley Johnson Hudson

**15:30-15:50** **Break**

**16:00-17:00** Identify Desirable Features of SNS Instrument

**18:00-19:30** **Dinner Chesapeakes**

**Saturday 2 February**

| | |
|---|---|
| **8:00-8:30** | **Continental Breakfast** |
| **8:30-9:00** | Recap of Friday Discuss requirements for Letter of Intent (LOI) |

**9:00-10:15** Breakup into Working groups
Science Case, Community Outreach, Instrument Design

**10:15-10:35** **Break**

**10:35-11:15** Regroup Reports from Breakout Group Leaders /
Discuss strategy / Assignments for LOI preparation

**12:00-12:45** **Lunch /Goodbyes**

**13:30** Tour SNS Construction Site Oak Ridge

## Fundamental Neutron Physics in the United States: An Opportunity in Nuclear, Particle, and Astrophysics for the Next Decade

Geoffrey Greene, Los Alamos National Laboratory,*
W. Michael Snow, Indiana University,
Christopher Gould, North Carolina State University,
Frank Plasil, Oak Ridge National Laboratory

### Executive Summary

Low-energy neutrons from reactor and spallation neutron sources are of great interest as experimental probes for the study of important questions in nuclear, particle, and astrophysics. While the primary focus of such sources are materials science studies through neutron scattering, there is a solid tradition of their productive and symbiotic use for nuclear and particle physics at facilities such as the Institut Laue Langevin, the National Institute of Standards and Technology Cold Neutron Research Facility, and the Los Alamos Neutron Science Center. There has been and continues to be an active and energetic United States community engaged in the area of research, including a number of excellent younger scientists. The scientific opportunities in this field include the elucidation of important issues in a number of areas, including

(1) the nature of time reversal non-invariance and the origin of the cosmological baryon asymmetry,

(2) the nature of the electroweak theory and the origin of parity violation,

(3) the nature and detailed description of the weak interaction between quarks,

(4) the origin of the heavy elements, and other issues in stellar astrophysics,

(5) the detailed investigation of quantum mechanics and precision measurements with neutron interferometry.

In each of these areas, there are specific opportunities that can best be addressed using a pulsed spallation neutron source. **The proposed Spallation Neutron Source at Oak Ridge National laboratory will provide the highest peak flux neutron source in the world and offers the United States scientific community an important opportunity in nuclear, particle, and astrophysics for the next decade**.

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*[*] The authors formed the organizing committee of the Workshop on Fundamental Physics with Pulsed Neutron Beams held in Triangle Park, NC, June 2000. The paper is intended to capture the consensus of opinion formed at the workshop. Notwithstanding this intention, any opinions expressed in text must ultimately be considered those of the authors.*

## Fundamental Neutron Physics in the United States: An Opportunity in Nuclear, Particle, and Astrophysics for the Next Decade

Geoffrey Greene, Los Alamos National Laboratory*
W. Michael Snow, Indiana University
Christopher Gould, North Carolina State University and TUNL
Frank Plasil, Oak Ridge National Laboratory

Low-energy neutrons from reactor and spallation neutron sources have been employed in a wide variety of investigations that shed light on important issues in nuclear, particle, and astrophysics; in the elucidation of quantum mechanics; in the determination of fundamental constants; and in the study of fundamental symmetry violation (Appendix A, Glossary). In many cases, these experiments provide important information that is not otherwise available from accelerator-based nuclear physics facilities or high energy accelerators. An energetic research community in the United States is engaged in "fundamental" neutron physics. With exciting recent results, the possibility of new and upgraded sources, and a number of new experimental ideas, there is an important opportunity for outstanding science in the next decade.

"Fundamental" neutron physics experiments are usually intensity limited. Researchers require the highest flux neutron sources available, which are either high-flux reactors (continuous sources) or spallation neutron sources (pulsed sources). The primary mission of these major facilities is neutron scattering for materials science research. Notwithstanding this condensed matter focus, essentially all neutron scattering facilities have accepted the value of an on-site fundamental physics program and have typically allocated 5 to10% of their capabilities (i.e., beam lines) toward nuclear and particle physics research activities.

Each experiment in a fundamental neutron physics program uses neutrons in a specific energy regime and a given experiment may or may not be well matched to the characteristics of a particular source. Experiments are distinguished by type of neutron beam that the facility must provide. See Appendix A, *Glossary*, for definitions of ultracold, cold, thermal, and epithermal neutrons, which will be used throughout the report. The neutron beams produced by high-flux reactors and spallation neutron sources differ significantly in their time structure. Particular experiments may be better suited to one source or the other.

Fundamental neutron physics has attracted an energetic community of academic and national laboratory researchers and includes a number of talented younger scientists. A special symposium at the October 1999 Division of Nuclear Physics meeting discussed nine different projects at the National Institute of Standards and Technology (NIST) and at the Los Alamos Neutron Science Center (LANSCE), involving ninety seven participants from twenty three different institutions (Appendix C, Extracts from the Bulletin of the American Physical Society, *Program for the Focused Session on Next Generation Fundamental Physics Experiments with Cold Neutrons*, Division of Nuclear Physics October 1999 Meeting). Activities in the fundamental neutron physics field have not been limited by a lack of interesting projects or a want of enthusiastic researchers but rather by a dearth of neutrons.

The broad range of scientific issues addressed by current experiments in fundamental neutron physics can be roughly placed into five categories as follows:

(1) the nature of time reversal non-invariance and the origin of the cosmological baryon asymmetry,

(2) the nature of the electroweak theory and the origin of parity violation,

(3) the nature and detailed description of the weak interaction between quarks,

(4) the origin of the heavy elements, and other issues in stellar astrophysics,

(5) the detailed investigation of quantum mechanics and precision measurements with neutron interferometry.
.

The first category, which lies at the heart of modern cosmology and particle physics, includes the search for the neutron electric dipole moment and the search for T-odd correlation coefficients in neutron beta decay ("D-coefficient" and "R-coefficient"). Among the important issues that are addressed by neutron experiments include whether or not the baryon asymmetry of the universe is directly related to fundamental T-violation and whether or not the magnitude of T-violation is consistent with the predictions of the Standard Model.

The second category involves the accurate determination of the parameters that describe neutron beta decay (lifetime and correlation coefficients). Comparison of these results can be used to see whether or not the weak interaction in the charged-current sector is completely left-handed (as it is in the Standard Model) or has right-handed components. These precision measurements can also provide important information regarding the completeness of the three family picture of the Standard Model through a test of the unitarity of the Cabibbo-Kobayashi-Maskawa (CKM) matrix. Neutron beta decay also provides the time scale for Big Bang nucleosynthesis and remains the largest uncertainty in cosmological models that predict the $^{4}$He abundance.

Category three involves the study of the weak interaction between quarks in the strangeness-conserving sector. This study is very difficult because of overwhelming direct effects of the strong interaction. As a result, the effective weak couplings in the usual meson-exchange model of the process are poorly known. In fact, different experiments yield contradictory results. Sensitive experiments using polarized cold neutrons to determine parity violation (an unambiguous tag for the weak interaction) in the n-p, n-D, and n-$^{4}$He systems provide an opportunity to measure NN weak interactions in simple systems that are not complicated by unknown nuclear structure effects. Knowledge of these interactions is required to understand parity violating phenomena in nuclei, such as the recently discovered nuclear anapole moment, and can be used to gain information on quantum chromodynamics (QCD) in the strongly interacting limit.

Category four examines stellar astrophysics and the origin of the heavy elements. Light element nucleosynthesis occurred during the first few minutes of the big bang; however, all isotopes with an atomic mass number greater than seven are created only in stellar processes. Typically, these stellar processes (“r, s, p, etc.”) involve competition between neutron capture, which moves isotopes to increasing atomic mass number and beta decay, which increases atomic number. The relative abundances are particularly sensitive to the neutron capture cross sections of radioactive nuclei with lifetimes comparable to s-process time scales (months to years). Intense neutron sources in the few keV energy regime (corresponding to stellar temperatures) provide the only experimental method of obtaining this information.

Neutron interferometry (category five), perhaps the most ideal realization of Schrodinger wave optics, has been employed to elucidate a number of phenomena in non-relativistic quantum mechanics. In addition, neutron interferometry is currently being used to perform precise scattering length measurements, which will eventually improve our knowledge of the electromagnetic structure of the neutron and address the question of nuclear three-body forces. Many important experiments have been suggested, but as the technique is extremely count-rate limited, only a subset of these have been performed. An intense pulsed source offers the possibility of extending these efforts into the study of time-dependent phenomena, opening up a range of new investigations.

Activities in fundamental nuclear physics are focused at a few high-flux facilities in Europe and the United States. The premier facility is the Institut Laue Langevin (ILL) in France, whose reactor is the highest flux continuous neutron source in the world. The ILL has maintained a vigorous program of fundamental neutron physics since the early 1970s. and has, in the past, been quite open to foreign scientists.

Researchers from the United States have been heavily involved in activities at the ILL since its inception and, indeed, have provided significant leadership in many major experiments. Now, however, there is considerable political pressure on ILL management to require some financial contribution to operations for United States experiments and future access to the ILL for United States researchers should not be taken for granted. The NIST Cold Neutron Research Reactor (with about one-third the power of the ILL) maintains three beam lines for fundamental neutron physics and is the current center of this activity in the United States. The LANSCE Manuel Lujan Jr., Neutron Scattering Center at Los Alamos National Laboratory is the highest peak flux neutron source in the world and supports a variety of nuclear physics activities. An aggressive program of cold and ultracold neutron experiments is in the planning stage at LANSCE.

It is very important to realize that the optimal neutron source for a given experiment will depend upon the details of that experiment. Essentially all the experiments in this program are significantly limited by both statistics and systematic effects. Achieving the highest statistical accuracy requires the highest flux source (i.e., reactors). However, the time structure of a less intense pulsed spallation source affords opportunities to examine systematic effects, which can outweigh the reduction in statistical sensitivity.

At least ten United States led fundamental neutron experiments have reached a sufficient level of maturity to be taken seriously in planning for the next decade (Appendix C, Extracts from the Bulletin of the American Physical Society, and Appendix D, Agenda for the International Workshop on Fundamental Physics with Pulsed Neutron Beams). These range from activities that have completed a first-phase measurement to speculative projects that will require extensive research and development before emerging as full-fledged proposals. A thoughtful national program in this area must be based upon a careful review of the scientific merits of each project, a determination of the most appropriate neutron source, and, of course, an assessment of the available resources. These resources include not only financial support but also the availability of neutron beams.

With the solidification of the Spallation Neutron Source (SNS) project and the interest by the National Science Foundation in a second, cold neutron, target station, the United States will have an extraordinary opportunity to develop leadership in fundamental neutron physics. United States researchers are now playing important roles in the development of superthermal ultracold neutron sources and in the elaboration of new ideas and techniques to exploit spallation neutron sources for fundamental physics experiments. The purpose of the recent June 2000 workshop (Appendix D, Agenda for the International Workshop on Fundamental Physics with Pulsed) was to identify whether or not any of these experiments would best be pursued at the SNS.

The conclusion was that a large fraction (perhaps one half) of the experiments discussed at the June 2000 workshop would indeed benefit from a spallation neutron source with the intensity and proposed moderator characteristics of the SNS. These experiments include, but are not necessarily limited to, the following categories (*brief comments on the advantages of a pulsed neutron source are included*):

(1) Experiments to measure the weak NN interaction, for example, gamma asymmetry in np and possibly nD capture, neutron spin rotation measurements in np and possibly nD and n-$^4$He (*A spallation neutron provides time-of-flight information which allows important checks of possible systematic effects*);

(2) In-beam neutron decay experiments that require absolute neutron polarization measurements (A and B coefficients) (*A spallation source provides neutron time-of-flight information which allows polarized $^3$He neutron polarizers to be exploited in a powerful way*);

(3) Neutron cross section measurements in the keV range on radioactive samples for nuclear astrophysics (*Only spallation sources produce neutrons in this energy regime. Neutron time of flight allows for the resolution nuclear resonances. The increased intensity of the SNS allows the study of interesting radioactive isotopes that are only available as very small samples*).

(4) Measurements with ultracold neutrons (such as neutron beta-decay measurements and the neutron electric dipole moment), which operate in a short-fill, long-counting mode. (*A high peak intensity pulsed source allows for greatly increased higher signal-to-noise ratios.)*

Although some of these experiments may be done at NIST or LANSCE before the SNS is operational, higher precision than is currently possible at these sources will be required to pursue the physics. In these cases, collaborators will be able to take an already-tested apparatus to the SNS. With many of the "bugs" already worked out of these experiments, the SNS should start to produce important physics results when the facility is turned on.

There is also a question as to whether or not other facilities in the world could compete with the SNS. Both the European Spallation Source and the National Laboratory for High Energy Physics (Japan)/Japan Atomic Energy Research Institute projects, which are both pulsed neutron sources, are still at the proposal development stage and have not yet been approved. A new cold neutron beam line for fundamental neutron physics has just been established at the ILL: this beam would be the most intense in the world and would certainly be the most appropriate beam for certain experiments, although as mentioned above its availability to United States researchers is uncertain. A fundamental neutron physics capability at the SNS will provide a totally unique facility and place the US in a position of unquestioned leadership in an important and exciting field.

We summarize briefly the main conclusion of the FPPNB 2000 workshop as follows:

***In each of the scientific areas in which fundamental neutron physics measurements have an important impact, there are experiments for which the SNS pulsed source is, without doubt, the source of choice.***

**Spallation Neutron Source Users Meeting**
Renaissance Hotel
999 9th St. NW
Washington, D.C.
**May 21-24, 2000**
Agenda

**Sunday, May 21, 2000**

1700 – 2100 Welcome Reception (Renaissance Ballroom East) and Registration (outside Renaissance Ballroom West A)

**Monday, May 22, 2000**

| | |
|---|---|
| 0700 – 0900 | Continental Breakfast (outside Renaissance Ballroom West A/B) |

General Session (Renaissance Ballroom West)

| | |
|---|---|
| 0900 – 0910 | Welcome (J. Decker, DOE) |
| 0910 – 0930 | Keynote Address (Under Secretary of Energy Ernie Moniz, DOE) |
| 0930 – 1030 | Impact of Neutron Scattering on Science (S. Sinha, ANL) |
| 1030 – 1100 | Break/Displays (outside Renaissance Ballroom, West A/B) |
| 1100 – 1200 | SNS Update and Overview (D. Moncton, SNS) |
| 1200 – 1400 | Lunch (Senator Bill Frist) (Renaissance Ballroom East) |

**Symposium on the Role of the SNS in Studies of Materials over the next 20 years**
Chaired by G. Aeppli, NEC Research

| | |
|---|---|
| 1400 – 1430 | Spallation Neutrons and Pressure: some exciting opportunities for condensed matter research (J. Parise, SUNY-SB) |
| 1430 – 1500 | Magnetism (S. Parkin, IBM) |
| 1500 – 1530 | Break/Displays (outside Renaissance Ballroom West A/B) |
| 1530 – 1600 | Soft Materials (M. Klein, U. Penn) |
| 1600 – 1630 | Engineering Materials (T. Gnaeupel-Herold, NIST) |
| 1630 – 1830 | NSSA Sponsored Reception (Congressional Hall A/B) |
| 1700 – 1830 | Introductions (J. Rhyne)<br>Remarks (Dr.Arthur Bienenstock, OSTP)<br>NSSA Forum (J. Rhyne) |

**Tuesday, May 23, 2000**

| | |
|---|---|
| 0700 – 0900 | Continental Breakfast (outside Renaissance Ballroom West A/B) |
| 0900 – 0930 | Introductory Remarks (Representative Zack Wamp)<br>(Renaissance Ballroom West) |

Update on the SNS Facility (Renaissance Ballroom West)
Chaired by J. Tranquada, BNL

| | |
|---|---|
| 0930 – 0955 | SNS Accelerator Complex (R. Kustom, SNS) |
| 0955 – 1005 | SNS Conventional Facilities (T. Chargin, SNS) |
| 1005 – 1030 | High Power Target Station (T. Mason, SNS) |
| 1030 – 1055 | Long Wavelength Target Station (J. Richardson, ANL) |
| 1055 – 1110 | Break (outside Renaissance Ballroom West A/B) |
| 1110 – 1500 | Parallel Breakout Sessions on SNS Instrumentation Priorities (working lunch – Renaissance Ballroom East)<br>Disordered Materials (M. Winokur, U. Wisconsin; H. Glyde, U. Delaware) (MR8)<br>Crystallography (A. Wilkinson, GA Tech; J. Jorgensen, ANL) (MR3)<br>Soft Materials (K. Blasie, U. Penn; R. Briber, U. Maryland) (Renaissance Ballroom West)<br>Magnetism (C. Broholm, Johns Hopkins; D. Argyriou, ANL) (MR-5)<br>Engineering Materials (S. Spooner, ORNL, X. Wang, SNS) (Auditorium) |
| 1500 – 1530 | Break (outside Renaissance Ballroom West A/B) |
| 1530 – 1630 | Reports from Breakout Sessions (Renaissance Ballroom West) |
| 1630 – 1700 | Status of the Long Wavelength Target Station Proposal (T. Mason, SNS)<br>(Renaissance Ballroom West) |

1830 – 1900 Cocktails before dinner (Grand Ballroom North)
1900 Conference Dinner (Grand Ballroom North)

**Wednesday, May 24, 2000**

0700 – 0900 Continental Breakfast (outside Renaissance Ballroom West A/B)

Symposium on Non-scattering Uses of the SNS (Auditorium)
Chaired by L. Schroeder, LBNL

| | |
|---|---|
| 0900 – 0910 | Introductory Remarks (T. Mason, SNS) |
| 0910 – 0935 | Opportunities for Neutrino Physics at SNS (F. Avignone, USC) |
| 0935 – 1000 | Nuclear Physics with Neutrons (G. Greene, LANL) |
| 1000 – 1030 | SNS Instrumentation and User Policies (T. Mason, SNS) |
| 1030 – 1045 | Break (outside Renaissance Ballroom West A/B) |
| 1045 – 1145 | Users Group Business Meeting (D. Belaner, UCSC) |
| 1145 – 1200 | Concluding Remarks (Congressional Hall B) |
| 1200 – 1330 | Lunch (Congressional Hall B) |

The following workshops are being held to discuss instrumentation and science at the SNS using long wavelength neutrons. These focused meetings are being funded by the National Science Foundation in conjunction with the Long Wavelength Target Station initiative:

Polymers, Colloids, and Biology at the University of Maryland, April 19, 2000

Magnetic Neutron Scattering at Argonne National Laboratory, April 27-28, 2000

Long Wavelength Powder Diffraction at Argonne National Laboratory, May 12, 2000

Liquids and Amorphous Materials at the University of Delaware, April 28-29, 2000

In addition, there are some other workshops that will also discuss science and instruments at the SNS that may feed into the LWTS process:

HP 2000: High Pressure Research at Argonne National Laboratory, April 7, 2000

Fundamental Physics with Pulsed Neutron Beams at the Research Triangle Park, North Carolina, June 1-3, 2000

# APPENDIX B

**Final Reports from Subawardees**

Subgrant No. OR11361-01.05, Final Technical Report

Title: Remote Data Access and Visualization System for LWTS Instruments.

**PROJECT PARTICIPANTS:** Dennis Mikkelson, University of Wisconsin-Stout.

**ACTIVITIES AND FINDINGS:**

1. Research and Education Activities:

This project developed a prototype remote data access system and designed a hierarchy of classes for neutron scattering data representation. This software was developed in collaboration with IPNS during the summer of 2001 and the 2001-2002 academic year. The work was done at the IPNS facility during the summer. During the academic year, the collaboration was continued via email, weekly tele-conferences and an additional on site visit.

2. Major Findings:

This section details the prototype remote live data access system and some of the ISAW infra structure improvements made under this project. Based on the experience developing these systems, some specific recommendations for further work are also made.

I. Remote Live Data Access

The data acquisition system at IPNS utilizes custom electronics modules that are controlled by a real time computer system running the VX-Works operating system. This part of the system resides in several VME crates. The real time control program is started from a separate "instrument computer". The instrument computer is a generic PC system running the Linux operating system. As histogram data is accumulated by the real time system, it writes its data to a file on an NFS mounted disk on the instrument computer.

A design priority for the live data server system was to not disrupt data collection in any way. The computing resources and software development tools available for the VX-Works systems are relatively limited compared to those available for the instrument computer. Consequently the live data server was designed to run on the instrument computer. The relationship of the systems is as shown in Figure. 1.

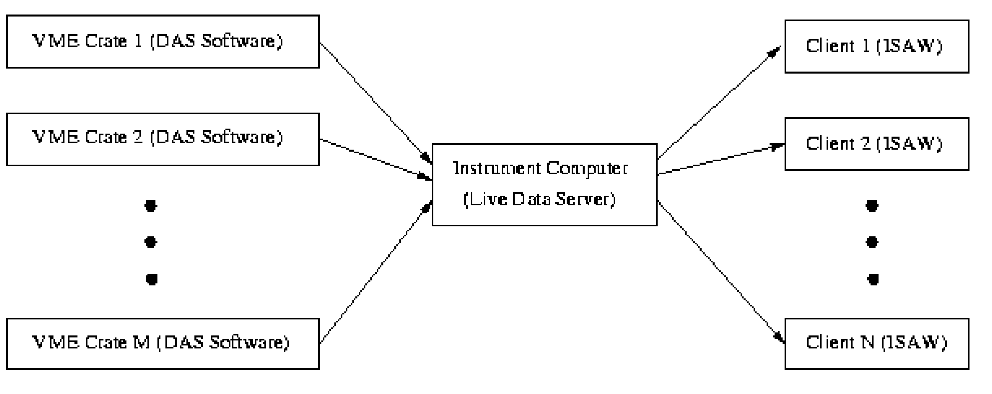


Figure 1. Live data access overview

The communication between the real time control computer and the live data server was kept as simple and reliable as possible. Specifically, the DAS software was modified to send UDP packets containing updated histogram data to the Live Data Server during the free time between pulses. UDP was used since UDP communication does not require handshaking. The UDP packets are sent by the DAS whether or not the Live Data Server is listening. Since the DAS never has to wait for the Live Data Server to acknowledge receipt of the UDP packets, the Live Data Server can't possibly disrupt the collection of data by the DAS. The UDP packets sent by the DAS have a very simple format as shown in Figure 2.

| | |
|---|---|
| 4 bytes | Magic Number |
| 1 byte | Instrument Name Length, n |
| n bytes | Instrument Name |
| 4 bytes | Run Number |
| 4 bytes | Group ID |
| 4 bytes | First Channel Index |
| 4 bytes | Num_channels, m |
| 4m bytes | Raw counts |

Figure 2

The DAS accumulates the full set of histograms for all detectors. As it sends the UDP packets to the instrument computer, it cycles through all of the histograms for all of the detectors. It splits the histograms into smaller pieces, as needed, so that each piece fits into a UDP packet.

The Live Data Server reads the structure of the current run from the empty runfile that is created when the run is started. This includes information such as the number of detectors, detector positions, number of bins in each histogram, etc. Using this structural information the Live Data Server can form a DataSet object and fill out the histogram values using the UDP packets from the DAS.

The communication between the Live Data Server and remote clients, which need the live data60, is done using TCP connections between the clients and servers. Java provides support for object "serialization". That is, objects can be "automatically" converted to a stream of bytes for storing in a file or for sending across a network. This provides an easy to use mechanism for sending complicated data structures to remote systems. The client-server system currently communicates by sending Java objects across the TCP connection. This is a very efficient way to handle the communication since the data is sent in large blocks without excessive overhead due to wasted time between transmissions of small data fragments. Currently, there is a simple command structure where the client requests a DataSet and the Live Data Server sends the requested DataSet.

In order to promote code reuse, the Live Data Server was structured to use the same lower level Java classes as a Remote File Server, also developed as part of the ISAW project. The major classes for the Live/File Data Servers are shown in Figure 3.

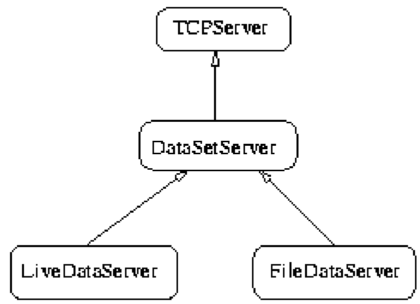

Figure 3. Server classes for remote data access

The responsibilities of each of these classes are:

TCPServer
- -Waits for requests from clients via TCP
- -Checks user name and password
- -Logs client requests

DataSetServer (extends TCPServer)
- -Defines commands for sending DataSets
- -Maintains list of data directories
- -Creates retriever for specified file ( .RUN or .NXS )

LiveDataServer (extends DataSetServer)
- -Receives UDP packets from data acquisition hardware
- -Forms DataSets from raw data
- -Sends DataSets to clients via TCP
- -Loads "empty" DataSets from initial run file
- -Switches to new run if UDP packet has new instrument or run

FileDataServer (extends DataSetServer)
- -Loads DataSets from file
- -Sends DataSets to clients via TCP

These classes use a number of simpler lower level convenience classes, UDPSend, UDPReceive, TCPComm, ThreadedTCPComm and TCPServiceInit, that handle the details of UDP and TCP communications in Java.

The client side of the communications is handled by the classes shown in Figure 4:

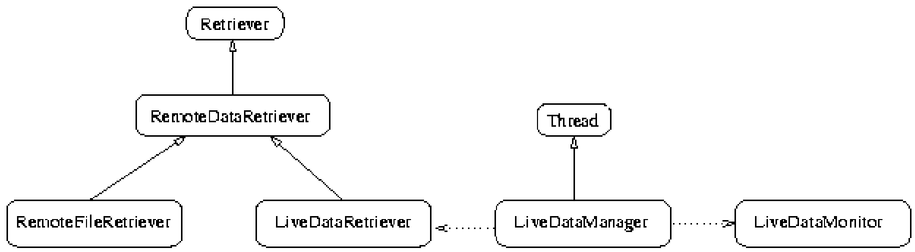


Figure 4. Client classes for remote data access

The responsibilities of each of these classes are:

Retriever (abstract base class for all DataSet retrievers)
-declares methods for getting the number and types of DataSets available from this source.
-declares method for getting a specific DataSet.

RemoteDataRetriever
-defines methods for making and breaking a connection with a remote data server
-defines method to get a specified object from a remote server
-defines error messages and method for getting the current state

RemoteFileRetriever
-defines the abstract Retriever methods for getting a DataSets from a specified file from a remote FileDataServer

LiveDataRetriever
-defines the abstract Retriever methods for getting DataSets from the current run on a remote LiveDataServer
-defines getStatus() method to determine the status of the DAS & LiveDataServer.

LiveDataManager
-periodically gets selected latest DataSets from a LiveDataRetriever
-periodically updates the LiveDataMonitor with the status of the DAS and LiveDataServer

LiveDataMonitor
-Displays status of LiveDataServer and DAS
-Displays list of available DataSets
-Allows user to select which DataSets are periodically updated

Live Data Server results and recommendations

There are at least two distinct uses for live data access that ultimately need to be addressed: instrument diagnostics and monitoring the experiment. These two areas require somewhat different information.

For instrument diagnostics the full monitor spectra may be needed to verify that the incident energy is correct. In addition, various types of reduced data such as the total counts for each detector together with some portion of the spectra at low energies, away from the elastic peak, could be useful to identify dead or noisy detectors.

For monitoring the experiment, properly summed and reduced data could allow relevant features in the data to be identified while the experiment is still running. This could in turn allow adjusting experimental conditions to optimize the use of beam time. Consequently, remote access to environmental data from the ancillary equipment is also needed.

The prototype LiveDataServer has been in fairly steady use on the High Resolution Medium Energy Chopper Spectrometer (HRMECS) at IPNS since August 2001. The primary use up to this point has been for instrument diagnostics. Currently, HRMECS has approximately 2000 detector elements. A connection to the Live Data Server can be made and a snapshot of the full set of histograms can be obtained in roughly 15 seconds.

This data rate is basically limited by the network bandwidth (100 Mbit/sec). Higher bandwidth networks are available and together with faster instrument computers higher data rates could be obtained. However, for instruments with one or two orders of magnitude more detectors, such as those planned for the SNS a more refined approach is needed. Specifically, full live access to all of the data being accumulated while an experiment is in progress will probably not be possible for larger instruments and may not be necessary. For many purposes, appropriately chosen subsets of the data and partially processed reduced data should be provided by the LiveDataServer.

Currently, several enhancements to the LiveDataServer are being planned. These enhancements will allow the client to specify which spectra or portions of spectra should be sent. The ability to request that the LiveDataServer first invoke some data reduction operators and then send the reduced DataSet is also being planned. Finally, since the ancillary equipment data can also be placed in a DataSet, this data can also be made available via the LiveDataServer.

As described in section 1, the prototype Live Data Server transmits the DataSets as serialized Java objects. The ability to serialize and easily send complicated data structures over the network greatly simplified the implementation of the prototype. Since the data is sent in large blocks this mode of communication is also quite efficient. However, there are two problems with this approach that will be addressed in future work.

First, the structure is quite inflexible. Specifically, as the system evolves, it may be necessary to add new objects or modify existing objects. Unfortunately, changing the data fields of an object, adding or removing methods or changing method signatures makes the modified data structure incompatible with the previous version. As a result, any such change requires that all clients be updated at the same time that the Live Data Server is updated. It has been possible to manage this for the limited deployment of the Live Data Server prototype, through careful planing of updates. However, this could potentially cause serious version conflict problems when deployed to a larger group of users at a large number of locations.

Second, communication via Java objects is Java specific. While the development of scientific software in Java is increasing, it is potentially useful to have access to the Live Data Server from programs written in languages other than Java.

There are several possible solutions to these problems. One possibility is to use CORBA. CORBA is freely available for many computing platforms and CORBA systems can be used from several different languages. Unfortunately, FORTRAN support for CORBA is questionable. The second possibility is to define a fairly simple protocol that would send the data as strings, numbers and arrays of numbers. To guarantee portability between systems, the data could be sent as ASCII text structured using XML. These options are being evaluated by the ISAW development team.

II. ISAW Data Representation

Neutron scattering data analysis is a computationally intensive process which often involves large quantities of data from multiple sources. While the largest volume of data is in the accumulated spectra (represented as histograms or collections of individual events), this data by itself is not meaningful. In order to be reduced and analyzed, "meta-data" such as detector positions, solid angles, incident energy, background measurements, sample temperature, pressure, etc. are needed. Different strategies for dealing with such data are possible. Traditionally, this data has been represented using simple data structures such as arrays. Unfortunately, this does not provide a connection between the accumulated spectra and the meta-data needed for reduction and analysis.

The ISAW project uses a flexible object-oriented approach to connect spectra with their meta-data. The two fundamental classes are Data and DataSet. A Data object corresponds to a single spectrum and a DataSet is a collection of Data objects. The meta-data corresponding to a particular spectrum is added to an extensible list of attributes for that Data object. The meta-data corresponding to a collection of Data objects is added to an extensible list of attributes for the DataSet object.

The Data object concept in ISAW was originally used to represent histograms. This project extended the Data object concept to explicitly include tabulated functions, model functions and histograms written in Java or defined by expressions, and histograms represented as a sequence of events. The new Data concept is the class hierarchy shown in Figure. 5

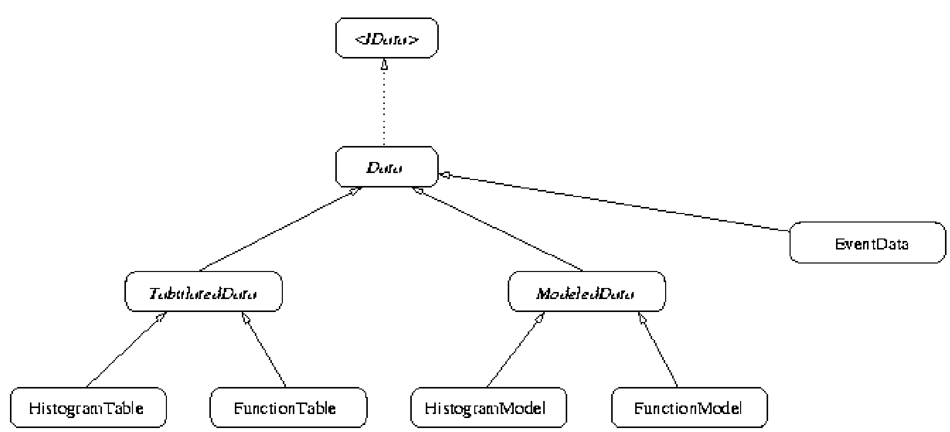

Figure 5. Data class hierarchy

The responsibilities of each of these classes are:

IData (interface specifying methods that any Data object must provide)
- -Declares method for extracting x,y and error values from a Data object, with or without smoothing.
- -Declares methods for arithmetic operations on the Data, add, subtract, multiply and divide using scalars or other Data blocks.
- -Declares methods for tracking group ID, selection, etc.
- -Declares methods for working with list of attributes and combining attributes
- -Declares method for determining whether or not a Data object represents a histogram

Data (abstract base class for all Data objects)

-Implements methods for arithmetic operations on the Data, using generic methods for extracting x,y and error values.
-Implements methods for tracking group ID, selection, etc.
-Implements methods for working with list of attributes and combining attributes.

TabulatedData (abstract base class for Data objects defined by tables of values)
-Implements methods for extracting x,y and error values from tables of values.

ModeledData (abstract base class for Data objects defined by Java functions and expressions)
-Provides constructors accepting functions of one varible describing the Data and error functions.

HistogramTable
-Provides constructors to build a HistogramTable from any Data block and from arrays of values. Constructing HistogramTable from function multiplies function(density) values times bin width to obtain frequency histogram.
-Provides method for rebinning histogram.

FunctionTable
-Provides constructors to build a FunctionTable from any Data block and from arrays of values. Constructing FunctionTable from histogram divides histogram values by bin width to obtain density function.
-Provides method for smoothing function.

HistogramModel
-Provides methods for evaluating defining function at specified x scale values.

FunctionModel
-Provides methods for evaluating defining function at bin centers of specified x scale values.

EventData
-Provides methods for building histogram values and errors from event data.

This class hierarchy allows experimentally sampled histograms and functions, modeled histograms and functions and event data to be operated on and viewed in the same way using the viewers and operators of ISAW.

The HistogramTable and FunctionTable classes are fairly conventional with their values defined by arrays of values. HistogramTable objects record the bin boundaries and counts for each bin. FunctionTable objects record function values at points.

The ModeledData classes have values that are defined by objects derived from the class OneVarFunction. See Figure 6. Derived classes include various parameterized functions that are useful for curve fitting, a class representing functions defined by a mathematical expression in a String, etc. These functions are evaluated at points as needed for viewing, or for performing operations.

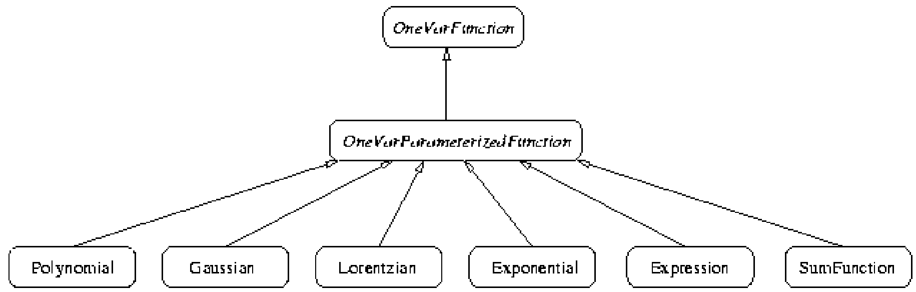


Figure 6. Classes for ModeledData and curve fitting

The responsibilities of some of these classes are:

OneVarFunction
- -Declares methods for evaluating function at single points and at arrays of points using single or double precision. Provides default implementations of these method, in terms of one abstract method for evaluating f(x) using double precision. Derived classes must provide at least that method.
- -Provides methods for tracking the domain of the function.
- -Provides method for numerical approximation to derivative.
- -Provides methods for handling name of the current function.

OneVarParameterizedFunction
- -Adds methods for dealing with a list of names parameters to a OneVarFunction.
- -Provides methods for numerical approximation of derivatives with respect to the parameters.

Polynomial
- -Represents a polynomial of arbitrary degree as a parameterized function with it's coefficients as parameters.

Gaussian
- -Represents a Gaussian peak shape with it's position, amplitude and full width at half max as parameters.

SumFunction
- -Combines an arbitrary sum of OneVarParameterizedFunctions into a new parameterized function.

The other classes in Figure 6, represent other commonly used functions..

The EventData object maintains a list of events in an EventList object. The events are binned into a histogram dynamically, when the histogram values are needed for viewing or operations. An EventList object is used for the low level event storage to allow for using different optimized representations for the list of events for different instruments.

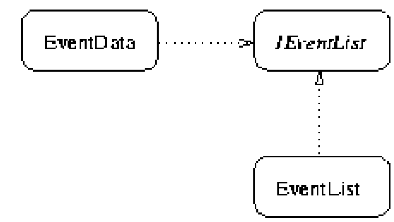


Figure 7. Classes representing event data

The responsibilities of each of these classes are:

EventData
-Extends the abstract Data class and uses a list of events to define histograms. The histograms are constructed dynamically from the events for any specified set of bins.

IEventList
-Declares a method for extracting a histogram as an array from a sequence of events.

EventList
-Stores an event list as a list of times where events occurred and the number of events that occurred at that time.

Currently there is only one class, EventList, that implements the IEventList interface. An EventList object records four pieces of information: a starting time, clock pulse width, a list of clock pulse numbers and a list of counts of events at that clock pulse. This data representation is actually a hybrid structure, part way between raw events and a histogram. However, for instruments such as direct geometry spectrometers, where most of the events are near the elastic peak, this provides a more compact form of the data than a complete histogram (including many zero bins) or a full event list with each event listed individually. The construction of a conventional histogram from this form of the EventList just requires stepping through the list of times where events occurred and adding the event count to the corresponding histogram bin. If the histogram being constructed has uniformly spaced time bin boundaries, this just takes O(N) where N is the number of times where events occurred. If the histogram being constructed has non-uniformly spaced bin boundaries, the construction of the histogram can be done in O(N log(M)) time, where M is the number of bins in the new histogram, using a binary search to find the correct bin. Alternatively, a merging algorithm could be used to construct the histogram in O(N + M) time. An adaptive algorithm could be made that would select the most efficient approach based on the characteristics of the data and the desired histogram bin structure.

This Data hierarchy allows sampled functions and histogram, modeled functions and histograms, and histograms recorded as sequences of events to viewed and operated on by the viewers, operators and scripts of ISAW in the same way. This use of the object oriented technique of polymorphism will allow event data from the SNS or other facilities to be usable in ISAW with very little additional work. However, due to the large data rates of the SNS instruments, further work on compression algorithms and data access for the EventList format for event data will be needed.

ACKNOWLEDGEMENT

The overall design of the Live Data Server was done by a group consisting of Alok Chatterjee, John Hammonds and Thomas Worlton from IPNS and Dennis Mikkelson and Ruth Mikkelson from the

University of Wisconsin-Stout. The modifications to the DAS Software to send UDP packets to the Live Data Server was done by John Hammonds. The code to access the ancillary equipment data files were provided by Alok Chatterjee, based on the SDDS software from the APS.

3.Opportunities for training and development.

4.Outreach activities.

**PUBLICATIONS AND PRODUCTS**

6.) The software developed for this project is incorporated in the ISAW system and is available from http://www.pns.anl.gov/sciandproj/isaw/.

2.) The Data hierarchy was described at the ACNS Conference, June 2002, Knoxville, Tennessee. A Flexible Representation for Time-of-Flight Neutron Scattering Data,
Mikkelson, D., Chatterjee, A., Hammonds, J., Loong, C.-K., Mikkelson, R., Peterson, P.F., Worlton, T.

**CONTRIBUTIONS:**

Remote access to experimental data, while the experiment is in progress will be a critical component at neutron scattering facilities as the user base for the facilities expands. The prototype live data server produced by this project is a first step in that direction. Currently it has been useful at the IPNS. The underlying software is NOT IPNS specific and can easily be adapted to other facilities. Several of the improvements to the remote data access system suggested in this report are currently being implemented, and we anticipate that it will also prove useful at other facilities. This will provide a freely available, portable mechanism for remote monitoring of experiments.

The EventData model developed as part of this project will allow event data from the SNS or other facilities to be accessed and processed seamlessly in ISAW.

**Contribution to final report for DMR-0073038 from Henry Glyde**

PARTICIPANT INDIVIDUALS

- Henry R. Glyde (CoPI) is a Professor of Physics at the University of Delaware. He has a research program on neutron scattering studies of Bose-Einstein condensation and excitations of quantum liquids in disorder.

- Jonathan L. DuBois is a senior PhD student at the University of Delaware who assisted in workshop organization, scientific information gathering, scientific case formulation and website preparation for the LWTS proposal. His research is on Monte Carlo simulation of Bose-Einstein Condensation in trapped Bose fluids.

ACTIVITIES AND FINDINGS

The essential activity was participation in the preparation of a pre-proposal and the full proposal for a Long Wavelength Target Station (LWTS) at the Spallation Neutron Source (SNS). This consisted of formulating the scientific case for the LWTS and associated neutron scattering instruments, setting specifications for the instruments, organization of meetings and workshops and writing sections of the proposals. Specifically, Henry Glyde:

- Organized the workshop at the University of Delaware with Dr. Lee Magid at which the plan for the preparing the pre-proposal to seek funds from NSF to write the full proposal was drafted.

- Reported to the Scientific Advisory Panel of the SNS at Argonne National Lab on the plan to write a pre-proposal and a full proposal for a LWTS.

- Attended subsequent meetings to prepare the pre-proposal.

- Wrote the section on “Liquids and Disordered Materials” of the pre-proposal.

- Held a two-day scientific workshop at the University of Delaware on “Liquids, Glasses and Disordered Materials” to engage the scientific community and set the scientific stage for writing the full proposal for the LWTS.

- Organized a scientific “Break Out” session on the LWTS at the 2000 SNS Users meeting in Washington, DC.

- Created a Website at UD on the above workshops and the science opened by the LWTS with a link to the SNS website.

- Attended many “Science and Instruments” meetings and meetings to prepare and discuss the proposal at ANL, Maryland and elsewhere.

- Wrote the section on “Liquids and Disordered Materials” and associated instruments of the LWTS proposal with Dr. Chun Leung of ANL

- Canvassed opinion of the scientific community on the LWTS proposal and provided some feedback on the proposal structure and content.

PUBLICATIONS AND PRODUCTS

1. Reports

a. Report on Workshop on Disordered Materials
Long Wavelength Target Station, Spallation Neutron Source
University of Delaware, April 28-29,2000

b. Report on Disordered Materials Parallel Breakout Session
SNS Users Meeting, Washington, May 21-24, 2000

2. Website

Created a website where above reports and others scientific LWTS-SNS materials appear : http://www.physics.udel.edu/conferences.html
http://www.physics.udel.edu/faculty/glyde

USE OF FUNDS

The funds provided were used for three purposes:

- Return of salary of HRG to the University of Delaware to obtain release from teaching for one semester to add above activity to schedule.
- To support a graduate student (Jonathan DuBois) for one year to assist in workshop organization, website preparation and scientific case formulation and information dissemination.
- To pay incidental expenses associated with workshops and some travel expenses not charged to UT.

OUTREACH ACTIVITIES

The chief outreach activities were holding scientific workshops and individual meetings with scientific community members to test and stimulate interest in the LWTS of the SNS as part of developing the scientific case for the LWTS. This included disseminating the scientific results of the workshops and creation of a website.

**Contribution to final report for DMR-0073038 from Angus Wilkinson**

## Project Participants

Prof. Angus P. Wilkinson was the only person at Georgia Tech who spent more than 160 hours working on the subcontract. However, some of the activities under the contract were performed in collaboration with Jim Jorgensen and Jim Richardson at the Argonne National Laboratory, Chicago, and several people from the US and the UK participated in a workshop that was used to help formulate plans for powder diffraction at the LWTS. The participants of the workshop are listed below.

## Activities and Findings

***Activities***: The subcontract to Georgia Tech was used to help formulate a plan for non-biological crystallography at the Long Wavelength Target Station of the SNS, help develop outline specifications for crystallographic instrumentation at the LWTS, and develop a strong scientific case in support of the planned instruments. Planning for crystallographic instrumentation at the LWTS grew out of a workshop (November 1998, Knoxville TN) organized by Jim Jorgensen on crystallography using neutrons at the SNS. In May 2000, a further workshop (Argonne National Lab) was run to explicitly consider both the scientific opportunities that could be realized using powder diffraction instruments on the LWTS and the types of instrumentation that would be needed to realize these opportunities. During the whole project period contact was maintained with other groups interested in diffraction instruments for the LWTS (in particular the molecular biologists) by attending planning meetings and workshops involving these groups. We also maintained contact with a broader community of potential users through participation in meetings such as the NSF sponsored Analytical Instrumentation for the New Millennium workshop in New Orleans (March 2001) and the NSF sponsored Future Directions in Solid State Chemistry workshop at U.C. Davis in Fall 2001. The PI on this LWTS subcontract also authored a section of the final report from the later workshop that discussed how national facilities such as the SNS might better serve the solid state chemistry community.

The May 2000 workshop started with an overview of anticipated LWTS performance from J. W. Richardson. This was followed by a report on the powder diffraction instrumentation for the HPTS from J. Hodges. Ken Anderson from ISIS UK provided a useful summary of his experiences with long-wavelength powder diffraction on the OSIRIS instrument at ISIS. Presentations covering some of the scientific opportunities in solid-state chemistry, molecular materials, mesoporous solids, nanoparticulate materials, in-situ catalytic studies, microporous materials, biomolecules and magnetism were made by K. Poepelmeier, P. W. Stephens, A. Stein, R. Whetten, J. Turner, B. Toby, R. VonDreele and D. Argyriou respectively. They were followed by a lively group discussion of scientific priorities and instrument characteristics.

***Findings:*** In general, the attendees at the May 2000 workshop believed that there were considerable opportunities in the areas of complex highly crystalline materials and materials displaying order on a length scale of a few nanometers. The possibility of studying organic materials in general and protein structures in particular by powder neutron diffraction received considerable discussion. While the examination of protein structure by powder diffraction methods (synchrotron X-ray) had recently been demonstrated to be highly effective for certain classes of problem, most attendees felt that it was too early to tell if powder neutron diffraction could have a big scientific impact on our understanding of protein structure.

The scientific opportunities that were uncovered broke down into two distinct groups: (1) Examination of ordered arrays of nanoparticles and ordered mesoposous solids, such as MCM-41. It was felt that this would ideally require an instrument capable of accessing d-spacings of up to 100 Å. (2) Complex large unit cell crystalline materials such as zeolites and many recently developed functional metal oxides. It is important to get high-resolution data over a wide d-spacing range if materials of this type are to be fully characterized by diffraction. The d-spacing range 0.3 – 40 Å was suggested as an appropriate target for a very versatile high-resolution medium flight path powder diffraction instrument on the LWTS and it was also thought that the low repetition rate of the LWTS would facilitate the construction of an ultra high-resolution instrument for examining very complex materials over a narrower d-spacing range. Such a long flight path instrument would offer the scattering contrast of a neutron experiment along with resolution that is currently only achieved on synchrotron powder diffractometers.

The scientific opportunities discussed seemed to dictate three different instruments: 1) a low resolution instrument spanning the d-spacing range 2 – 100 Å, that would probably be best developed along with the small angle scattering community; 2) a high resolution medium flight path instrument capable of covering the d-spacing range 0.3 – 40 Å, that would also probably be of interest to the magnetic materials community; 3) an ultra high-resolution long flight path instrument perhaps covering the d-spacing range 0.3 – 4 Å. These instruments distinguish themselves from what is possible on the HPTS by offering access to an extended d-spacing range and superior resolution. Where count rate is as at a premium and d-spacing range/resolution are secondary considerations, for example during parametric studies, the instruments on the HPTS would clearly be superior.

On the basis of the discussions at this workshop, a more detailed scientific case for the construction of two powder diffraction instruments at the LWTS was developed and incorporated into the LWTS proposal. Additionally, it was suggested to the small angle scattering community that they should consider building an instrument that had good capabilities in the 2 –100 Å d-spacing range. The scientific case for these instruments was primarily written at Georgia Tech with advise from many people experienced in powder diffraction. The final scientific case emphasized very strongly the scientific impact that an ultrahigh resolution instrument could have on the characterization of very complex materials. The LWTS offered an opportunity to build a neutron diffractometer capable of recording a diffraction pattern with synchrotron like resolution using modest sample sizes in reasonable amounts of time. For many applications these data sets would be vastly superior in information content to what could be obtained using a synchrotron (high Q data with good signal to noise, sensitivity to magnetism and light elements) and in very difficult cases a combination of neutron data from the proposed instrument and synchrotron data would advance considerably the complexity of problem that could be solved using powder diffraction.

***Education and training***: The construction of the proposed instruments would have affected the thesis research of many graduate students. Both of the proposed instruments were capable of high sample throughput and were likely to have a broad user base as powder diffraction is applicable to many different classes of materials. Currently, the areas under study using powder diffraction (X-rays as well as neutrons) range from proteins, fundamental materials physics and chemistry, earth science, designer porous solids and self assembled nano-structures through to engineering alloys and cement. The development of new instruments and capabilities would have further spurred the growth of both the depth and breadth of the user community.

***Outreach activities***: There were no outreach activities as part of the subcontract to Georgia Tech.

## Publications and Products

The major products of the subcontract were (1) a recommendation to build two powder diffractometers at the LWTS, an ultrahigh resolution instrument and a high resolution wide d-spacing range instrument, and (2) the scientific case for powder diffraction that was ultimately included in the LWTS proposal.

## Contributions

The proposed powder diffraction instrumentation for the LWTS, if built, would have enabled many previously impossible structural studies of materials that are of current interest to the materials physics and chemistry, earth science, and materials science communities. The instruments would have had a direct impact on the education and training of many graduate students in these communities and, through the proposed LWTS/SNS outreach programs, it would also have had an impact on undergraduate and K-12 students. As many of the materials to be studied using the proposed powder diffraction instruments are of technological relevance, for example microporous catalysts, ferroelectrics and magnetic materials, it is reasonable to expect that some of the work done on these instruments would have led to economic benefits and improvements in the quality of life for individual citizens.

**Final Report, Subaward to the University of Illinois at Urbana-Champaign, Brent Heuser, PI**

The motivation for this component of the project was to determine the utility of a pelletized moderator. Such a moderator allows solid methane to be used at higher target power levels lads since flowing liquid hydrogen provides the necessary heat removal. However, it is important to determine if critical neutron leakage characteristics—pulse width and leakage intensity—are adequate in the pelletized case. The simulations presented below demonstrate that a pelletized moderator has performance at least as good as a liquid hydrogen moderator and, in one case, performs better than liquid hydrogen.

Cold moderator performance has been investigated at the University of Illinois using the neutron transport simulation code MCNPX. A target-moderator geometry consisting of three cold moderators; two coupled (one slab, one wing) on the starboard side and one decoupled slab on the port side. The primary purpose of these simulations is to determine the effectiveness of a mixed, heterogeneous moderator consisting of solid methane pellets embedded in liquid hydrogen. Early in our work it was discovered that a homogeneous moderator (that is, without pellets) with the same composition of a pelletized moderator (for a given packing fraction) gave identical results within statistical error. Since the computation time is was greatly reduced for the homogenized case, all subsequent simulations were performed using homogenized moderators.

Figures 1-3 show the neutron leakage spectra from the three moderators for three different materials: solid methane (includes 10% by volume aluminum sponge), liquid hydrogen, and homogenized (corresponding to a pellet-liquid hydrogen packing fraction of 48%). Notice that the spectra for different moderator materials in a given moderator are not equal in the higher energy 1/E range. We believe this effect is due to attenuation in the methane-containing materials, which have a higher hydrogen density. To check this the thickness of the moderators were adjusted by the ratio of the hydrogen number density. The result of this optimization procedure on the starboard slab moderator is shown in Figure 4. Notice that the spectra are approximately equal in the 1/E energy range. This thickness optimization procedure yielded similar results for the other two moderators. These results are not shown.

At this point it is useful to examine the ratio of the leakage tally versus energy for the different cases. Figures 5-7 show the tally ratios for the three moderators without thickness optimization normalized to the liquid hydrogen case. The same ratios for the thickness-optimized port and starboard slab moderators, again normalized to liquid hydrogen, are shown in Figures 8-9.

The effect of pellet packing fraction was also investigated. A random, non-periodic distribution of spheres has an ideal packing fraction of approximately 65%. The 48% packing fraction used in the simulations presented above is significantly below this value. The tally ratio for packing fractions of zero (no methane pellets), 60%, and 70%, all normalized to the 48% tally, are shown in Figure 10 for the starboard slab moderator. These results were obtained with a homogenized system. This figure demonstrates that packing fractions from approximately 50 to 70% give nearly identical results above a few Angstroms.

The results thus far have focussed on spectral intensity. The other important moderator characteristic is pulse width in the time domain. Plots of intensity versus time for a narrow wavelength range are not too interesting when taken alone. More relevant is the combination of neutron leakage intensity and pulse width. In fact, a common figure of merit (FoM) is the ratio of leakage intensity to the square of the pulse FWHM, $I/FWHM^2$, versus neutron wavelength. FoM values for all three moderators are shown in Figures 11-13. In all cases, the solid methane materials has slightly better performance at shorter wavelengths, at least based on this FoM. The homogenized material has a performance between the solid methane and liquid hydrogen at shorter wavelengths. Smaller differences in the performance of the three materials are observed at the long wavelengths for the starboard wing and port slab moderators. Noticeable differences in the homogenized case compared to the liquid hydrogen case are evident in the starboard slab moderator.

Taken together, these results indicate that a pelletized cold moderator does not significantly enhance performance. Clearly the intensity at long neutron wavelengths are enhance relative to liquid hydrogen for decoupled moderators (Figure 6, starboard wing). A coupled liquid hydrogen moderator appears to give a higher yield at the peak leakage intensity (Figure 1, port slab). More importantly, the FoM values, which preferentially weight pulse width, indicate that long wavelength performance is comparable for all three moderator materials. The performance of the three moderator materials do diverge near the spectral peaks. However, cold moderators are typically used in long wavelength applications; a liquid hydrogen moderator is probably the best choice if the use of solid methane is prohibited.

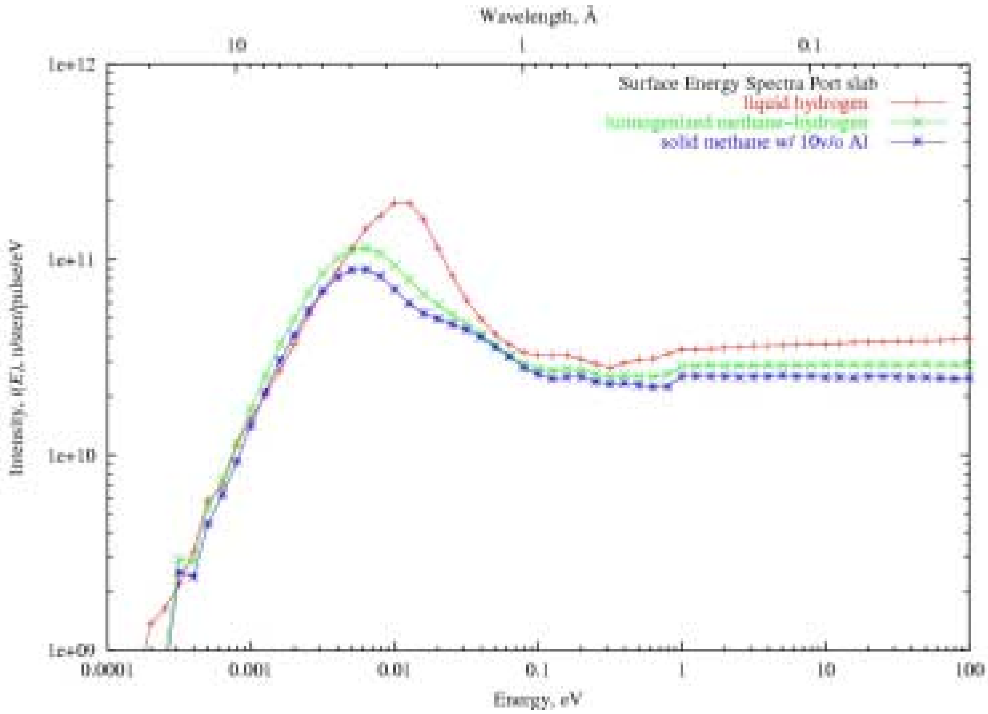


Figure 1. Leakage spectra for port slab moderator. This moderator is coupled.

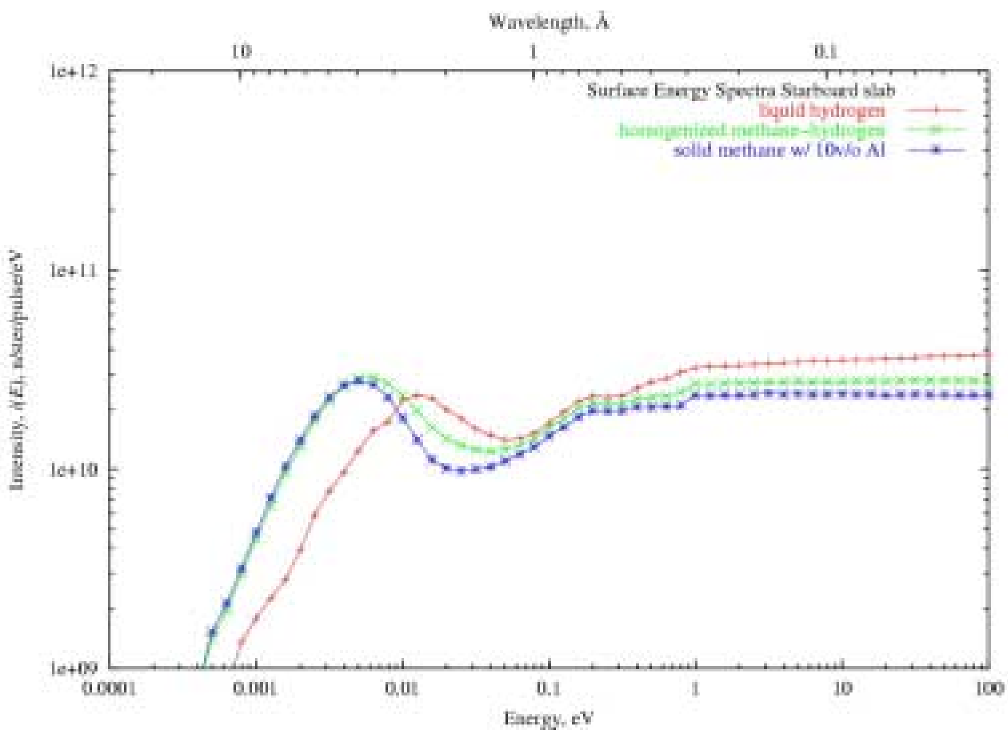


Figure 2. Leakage spectra for starboard slab moderator. This moderator is decoupled.

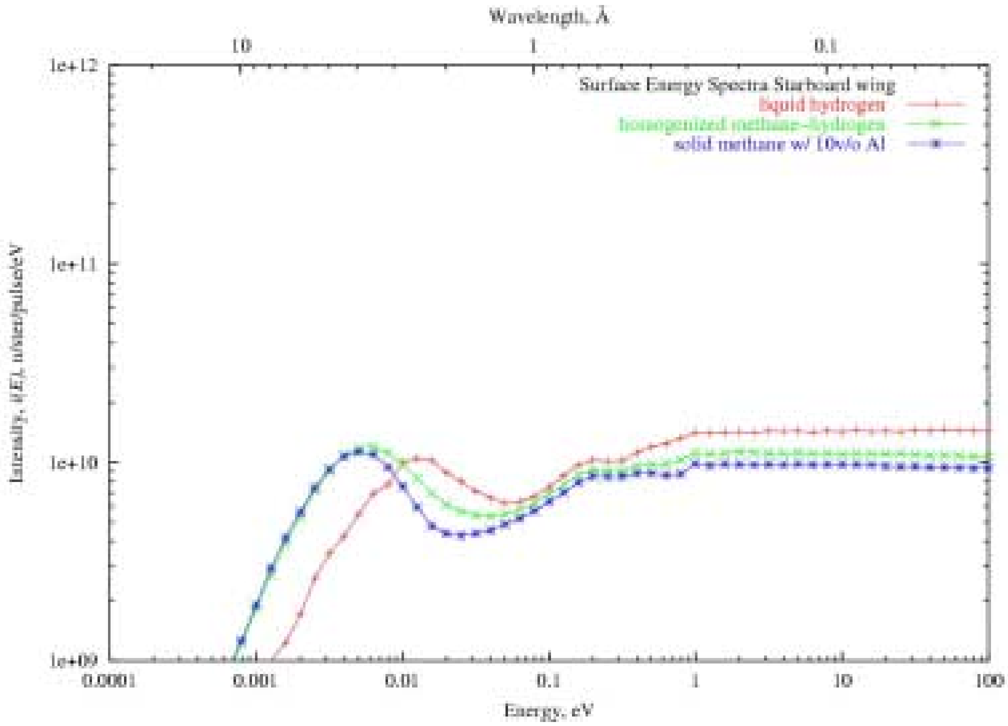


Figure 3. Leakage spectra for starboard wing moderator. This moderator is decoupled.

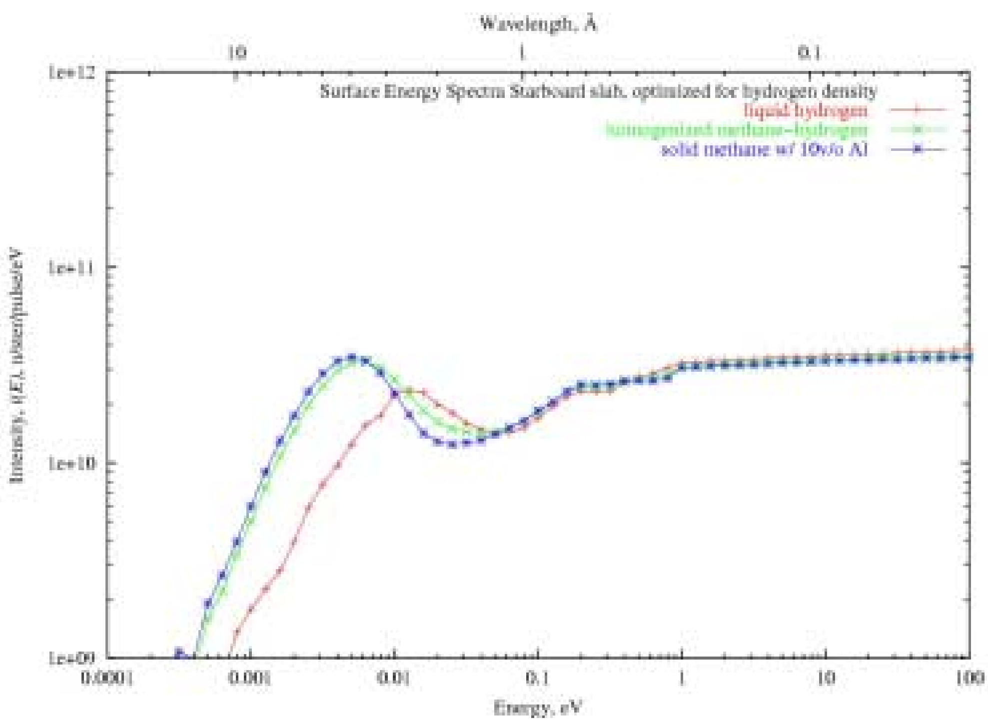


Figure 4. Leakage spectra for the starboard slab moderator showing effect of optimizing the moderator thickness by the hydrogen number density.

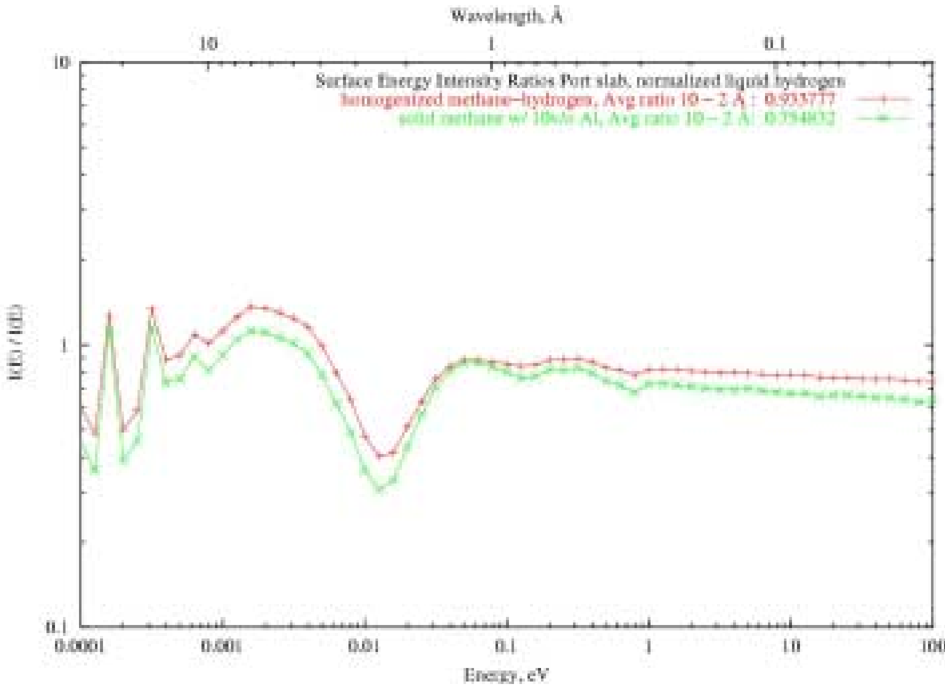


Figure 5. Tally ratio for the port slab moderator, without thickness optimization.

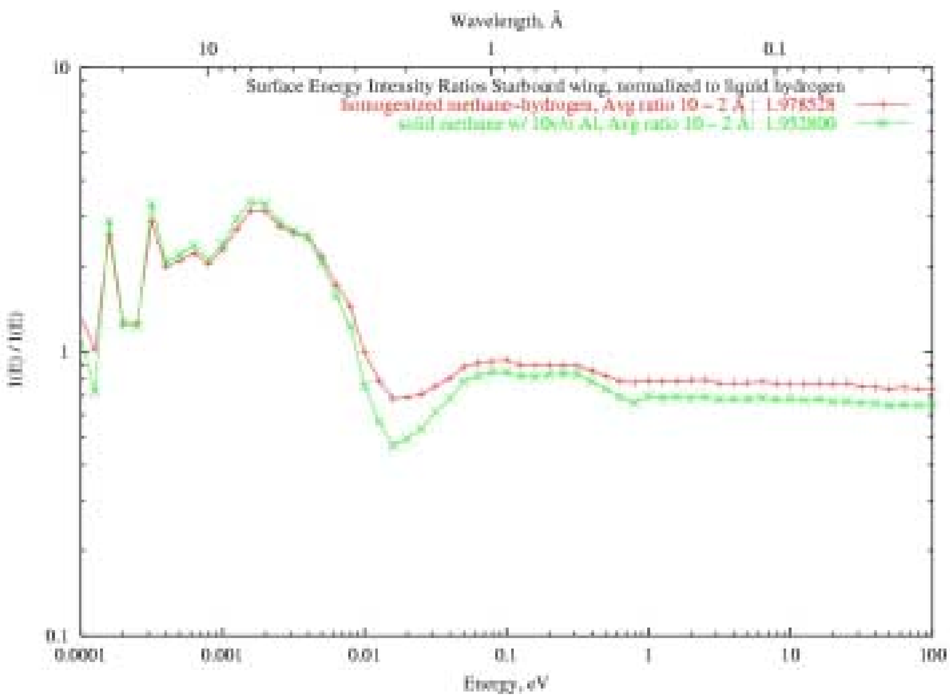


Figure 6. Tally ratio for the starboard wing moderator, without thickness optimization.

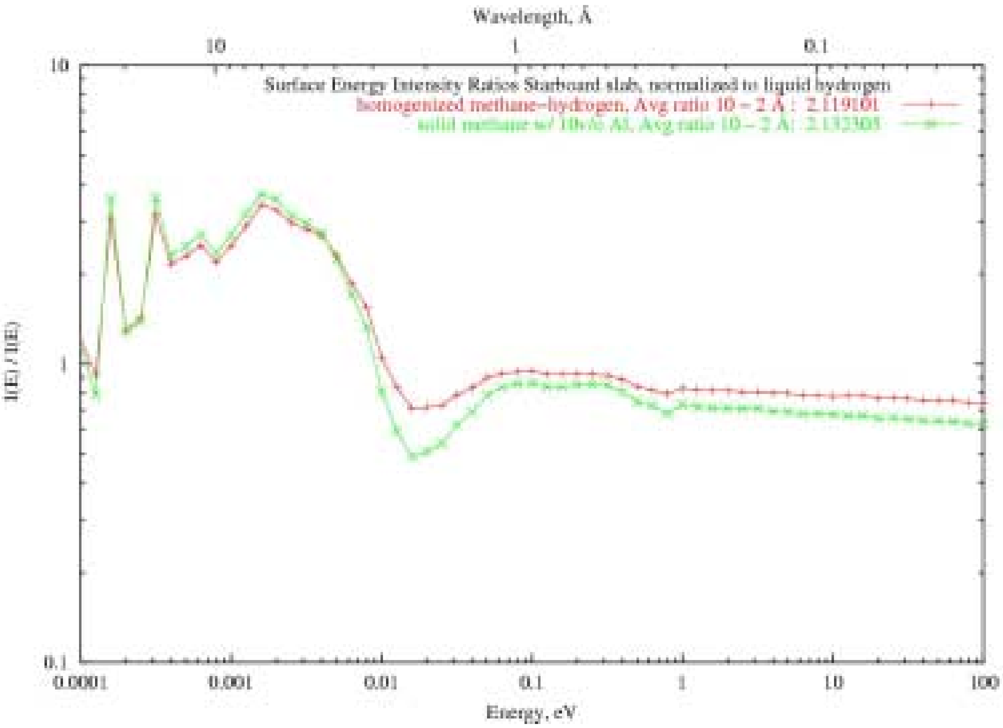


Figure 7. Tally ratio for the starboard slab moderator, without thickness optimization.

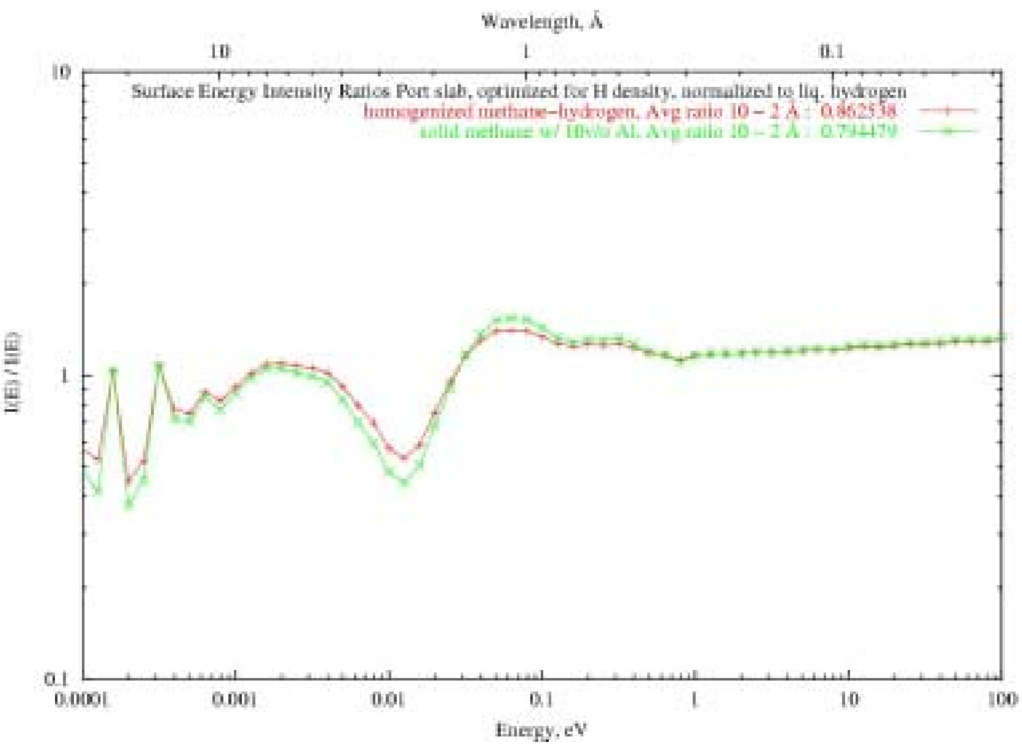


Figure 8. Tally ratio for the port slab moderator, with thickness optimization.

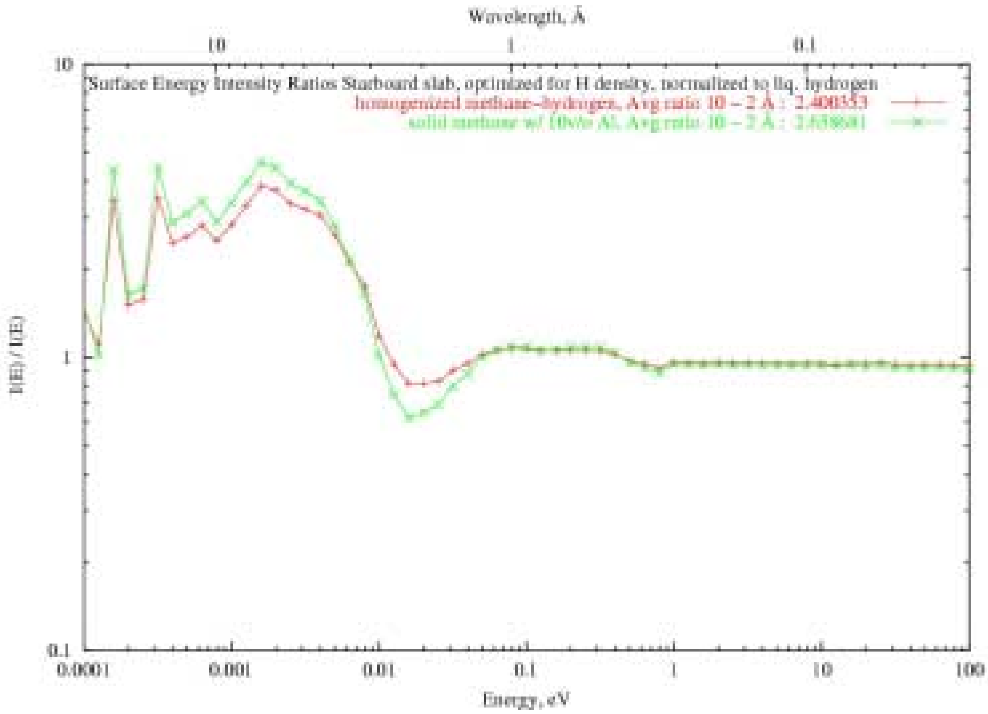


Figure 9. Tally ratio for the starboard slab moderator, with thickness optimization.

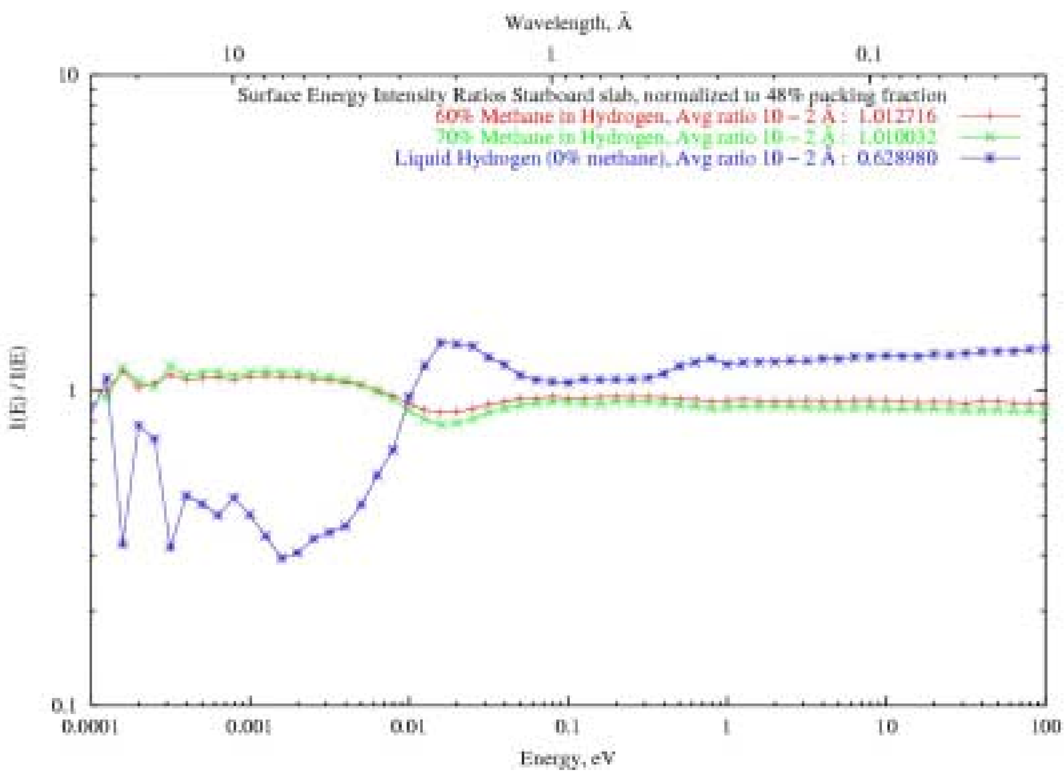


Figure 10. Effect of pellet packing fraction on tally ratio, normalized to the 48% packing fraction for the starboard slab moderator.

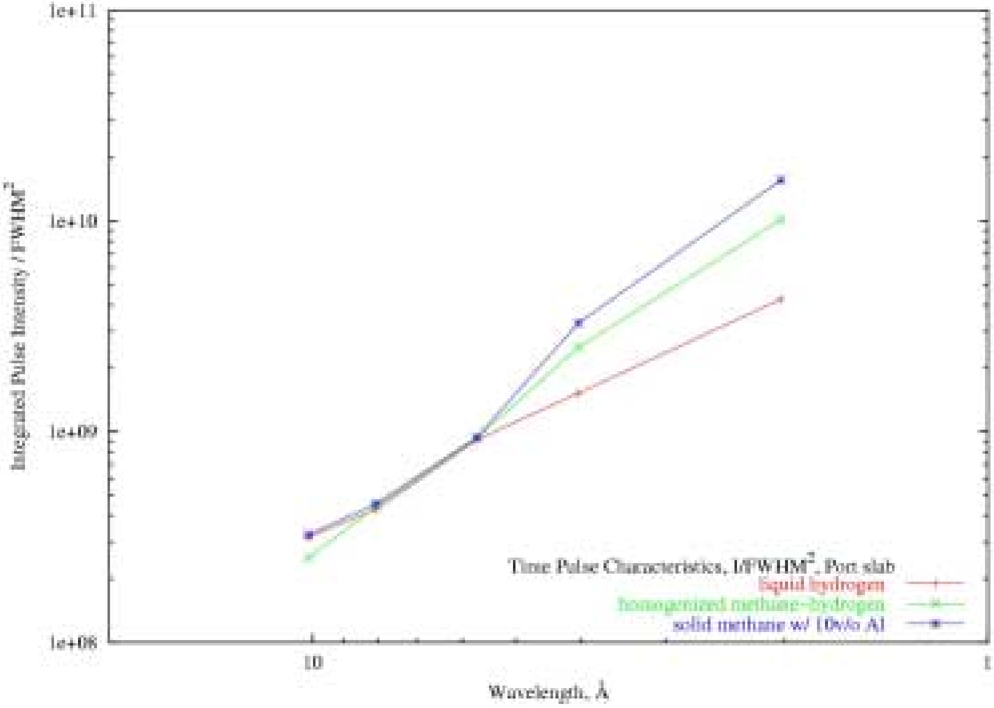


Figure 11.  Figure of merit of the port slab moderator for the different materials.

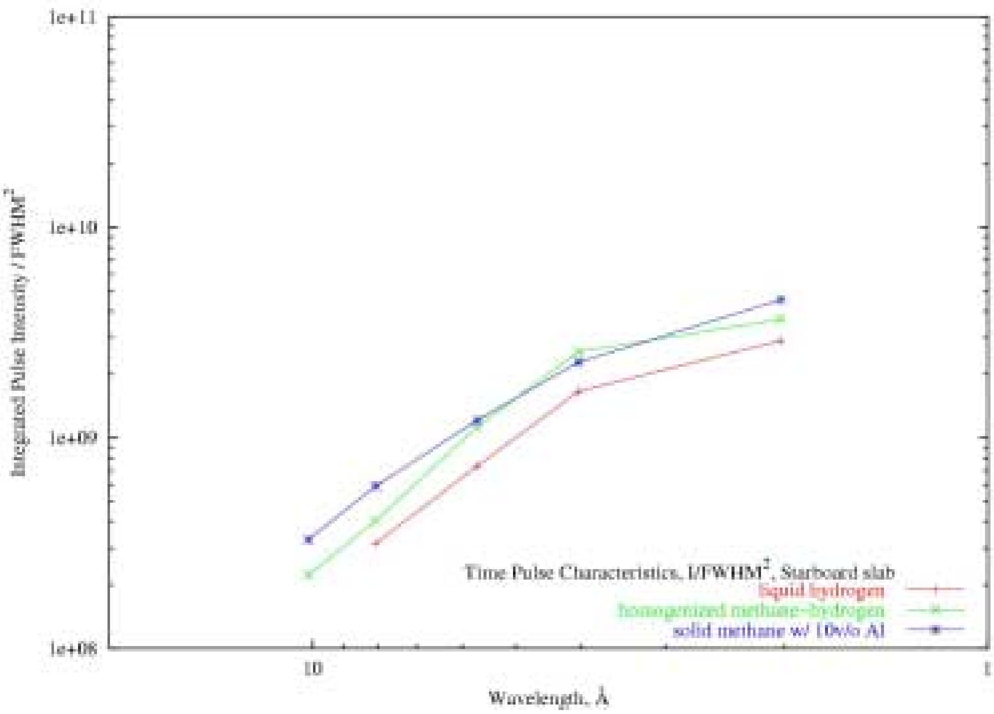


Figure 12.  Figure of merit of the starboard slab moderator for the different materials.

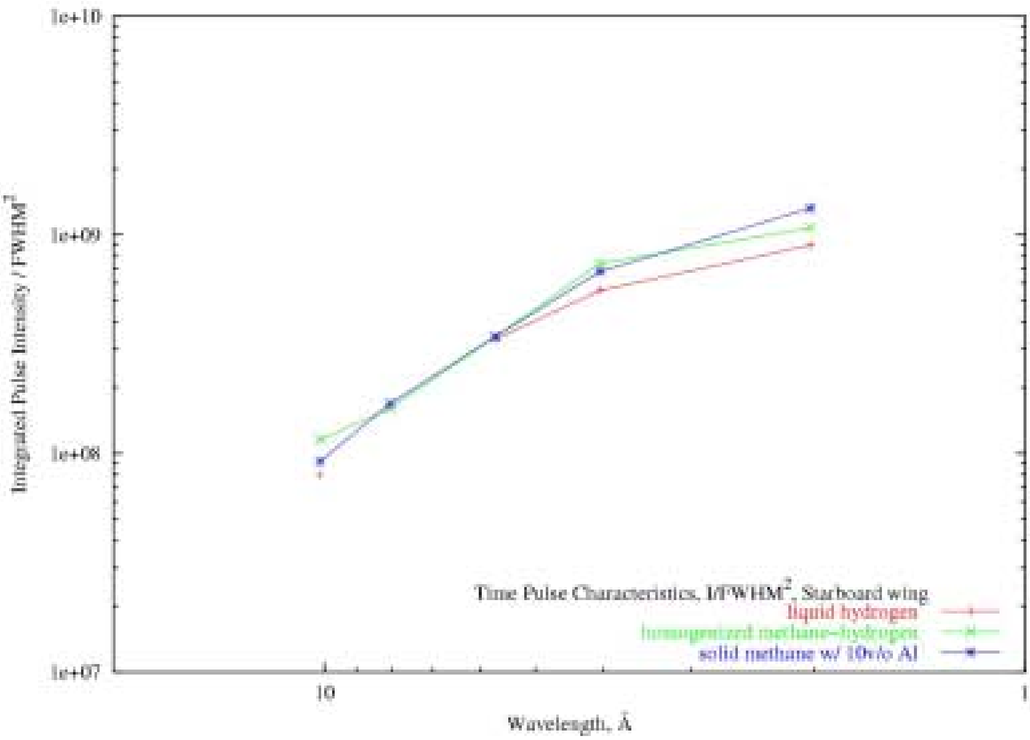


Figure 13. Figure of merit of the starboard wing moderator for the different materials.

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