A report on the

SNS Inelastic Neutron Scattering Workshop

held November 1, 1999 at
Argonne National Laboratory

SNS Document Number IS-1.1.8.2-8004-MM-A-00
January 31, 2000

Workshop organizers: D. Abernathy
S. Kern

Report compiled by: R. McQueeney
S. Nagler
P. Sokol
F. Trouw

Contents

A group of researchers representing several disciplines met at Argonne National Laboratory to establish scientific priorities for the inelastic neutron scattering instruments to be available at the SNS at the beginning of operations. The Workshop consisted of breakout sessions into three groups representative of different user communities for inelastic scattering: i) magnetism, ii) thermodynamics, lattice effects, and critical phenomena, and iii) chemical spectroscopy. A joint meeting of all three groups to discuss common interests and explore the overall needs for instrumentation followed. This report summarizes these discussions.

The report is organized as follows: Section I enumerates the instruments considered for inclusion in the Inelastic Scattering suite of instruments for SNS. Sections II(A,B,C) report on the scientific needs identified in the breakout sessions. Section III consists of a summary and the recommendations of the working groups.

I. Instruments

The instruments considered were broken up into three categories as summarized in table I below. Category A consists of instruments already identified by the SNS Instrument Oversight Committee as high priorities for study and design. Additional instruments, denoted by category B, have been identified for priority consideration at this Workshop. Other instruments which also should be discussed are in category C. The instruments in Table I are labeled by category letters and numbers. The numbers are for convenience of discussion only and do not reflect priorities. Further details of the instruments are discussed in sections II and III.

Table I : Instruments Discussed

Category	Instrument
A.1	indirect geometry spectrometer, (backscattering), resolution 2 μeV (elastic position)
A.2	Direct geometry spectrometer (chopper), resolution $\Delta E/E \sim 1\%$ (elastic position), E ~10 – 1000 meV, continuous angular coverage
A.3	Spectrometer with 10 – 100 μeV resolution
B.1	Chopper , $\Delta E/E \sim 5\%$ (elastic position), large angular coverage
B.2	Inverse geometry spectrometer, time focussed, $\Delta E/E \sim 1\%$
B.3	Triple axis-like instrument with high signal to noise
B.4	High Q chopper spectrometer with small $\Delta E/E$
C.1	Spin echo spectrometer $\Delta E \sim$ 1 neV to 2 μeV
C.2	Brillouin scattering spectrometer, small Q, intermediate E
C.3	PRISMA-like spectrometer

II. Scientific needs

IIA. Magnetism

Participants:

John Ankner, ORNL/ANL (recording secretary)
Ward Beyermann, U. of California at Riverside
Eugene Goremychkin, ANL, JINR
Sandy Kern, Colorado State. U./ANL
Thom Mason, ORNL
Ray Osborn, ANL
Roger Pynn, LANL
Jim Rhyne, U. of Missouri
Sunil Sinha, ANL
John Tranquada, BNL
Stephen Nagler, ORNL (chair)

Discussions:

The breakout session discussed the current scientific frontiers related to the dynamics of magnetic materials and the type of neutron instrumentation needed to address them. Examples of problems for which the study of magnetic excitations is essential include:

- Crystal field levels and low frequency magnetic fluctuations in correlated electron systems, including rare earth, heavy fermion, and non-fermi liquid ground state materials
- high temperature superconductors and related materials
- spin density wave materials
- colossal magnetoresistant materials, including manganites
- amorphous magnetic materials
- magnetoelastic effects in superconductors, e.g., borocarbides
- random and frustrated magnetic systems
- low dimensional and quantum magnetic systems
- spin Peierls and other spin gap materials
- magnetic films, multilayers and nanoparticles
- molecular magnets

A critical element that emerged from the discussions is that all instruments should be designed from the outset to accommodate a broad range of sample environment equipment. For most systems of interest, it is essential to study samples as a function of temperature, which in different cases can vary from milli-Kelvin temperatures obtained using dilution refrigerator techniques to temperatures greater than 1000 K requiring specialized furnaces. Measurements in the presence of externally applied magnetic fields are often crucial, and it may be anticipated that steady state fields ranging up to nearly 20 Tesla and pulsed fields at higher values may be practical by the time SNS is fully operational. In some instances controlled high pressure environments variable up to several GPa are desirable.

Another element common for all instruments is the desirability of increasing the neutron intensity incident on the sample via neutron optical techniques. Frequently, only very small samples of the most interesting new materials are available. The possible use of convergent guides, focussing monochromators, and compound refractive lenses should be considered carefully during the design phase of each inelastic instrument. Finally, the capability of using neutron polarization analysis would be extremely helpful for all inelastic magnetic scattering instruments. The possibility that real time measurement of the polarization dependence of the scattering could be used in some fashion was also raised.

Various types of inelastic scattering instruments were identified as potentially useful for inelastic magnetic scattering. The magnetic form factor in the scattering cross-section dictates that most magnetic scattering of interest is measured at relatively small values of momentum transfer ($0 < Q < 6$ Å^{-1}); nevertheless measurements at large momentum transfer are necessary to determine the background arising from non-magnetic scattering. The continued growth in the use of pulsed neutron methods as a primary tool for investigating magnetic excitations in single crystals implies that vector **Q** will be measured in many experiments. The requirements for energy transfer and resolution vary widely. For this reason a variety of instruments is required to meet the needs of the research community.

The magnetism working group at the November 1998 workshop identified four instruments as having the highest priorities for initial operation at the SNS. The present workshop participants were, for the most part, in substantial agreement with those recommendations. However, some new instruments were suggested and there was a good deal of discussion about the merits of more innovative instrument concepts. The instruments discussed in 1998 correspond roughly to A1, A2, A3 and B1 listed in Table I. Significant interest was also expressed in the instrument B3.

To elucidate the physics of small spin-gap systems, ground state splittings and low lying crystal field levels, and quasi-elastic scattering, a high resolution indirect geometry spectrometer (A1) was felt to be very important. Compared to existing instruments, there is a need for greatly reduced background and elimination of spurious scattering to maximize the impact this instrument would have on magnetism-related research.

For many magnetic systems, most of the physics occurs at energies less than 25 meV, and an energy resolution of 100 μeV is perfectly adequate. This is provided by instrument A3. This instrument was identified as an inverse geometry spectrometer at the November 1998 workshop, but the current consensus is that the precise design is not yet well determined.

Measurements on single crystals with dispersive magnetic excitations demand continuous coverage of Q with reasonably good Q resolution and flexible E resolution. The spectrometers A3 and B1 are seen as workhorses that could drive innovative research into new materials and forefront problems with wide variability in characteristic excitation energies. These should be in constant high demand.

It did not escape notice that the time averaged flux at the SNS will approach that available at a medium flux reactor. This opens up the possibility of utilizing triple-axis like methods to examine details of excitations when the important physics can be found at specific, well defined regions of **Q** and E space. A spallation source instrument can have significant advantages because the time structure allows for a tremendous reduction in background and possible rejection of the spurious scattering processes that plague triple-axis measurements at steady-state sources. This possibility is exciting and warrants serious consideration.

IIB. Thermodynamics, Lattice Effects, and Critical Phenomena

Participants:

Doug Abernathy, ORNL/ANL (recording secretary)
Ken Andersen, ISIS
Feri Mezei, LANL/HMI
Rob McQueeney, LANL
Paul Sokol, Penn State U. (chair)

Discussions:

The breakout session on thermodynamics, lattice effects, and critical phenomena discussed many topics covering a wide range of scientific disciplines. The topics were discussed mainly according to more general physical phenomena, rather than specific systems:

- Quantum fluids
  - Excitations at low Q
  - Collective and single particle excitations
- Phonons
  - Soft modes
  - Electron-phonon coupling
  - Phonon dispersions
  - Phonon densities-of-states
  - Small polarons (short-ranged lattice excitations).
- Disordered and amorphous materials
  - Relaxations
  - Glass transition
  - Excitations
- Momentum distributions
  - Hydrogenous and other systems
  - Anisotropic bonding

- Diffusion
    - H in metals
- Liquids
    - Diffusion
    - Low Q excitations
- Low dimensional systems

In addition to the necessary accommodation of a wide range of sample environment, optical and polarization equipment, as stated in summaries of the other breakout sessions, the group also discussed the need for state-of-the art data acquisition and analysis equipment and software. Especially in the measurement of single crystal samples, sampling vector **Q** on most pulsed source instruments implies a large effort for data handling, visualization, reduction and analysis. Resolution effects need to be well understood and software optimized for performing convolution/deconvolution of data. These considerations will likely entail the use of advanced programs for experimental planning and visualization; this software should be made readily available to outside users.

Due to the wide array of scientific disciplines covered in this breakout group, there were correspondingly many demands on inelastic scattering instrumentation. The instrument needs of this group spanned orders of magnitude in both energy and momentum transfer and their corresponding resolutions. For example, measurements of momentum distributions require good epithermal fluxes (incident energies up to 1 eV), large momentum transfers (up to 50 $Å^{-1}$) and reasonably good resolutions ($\Delta E/E_i \sim 1\%$, $\Delta Q/Q \sim 0.5\%$), whereas relaxation phenomena in glasses require cold neutrons (1-10meV), momentum transfers down to $0.1Å^{-1}$ and energy resolution ranging from neV to 100 μeV.

Many of the topics discussed in this breakout group did share a common feature centered around the study of short-ranged excitations (examples being liquids, glasses, alloys, impurity states and polarons). The impurity problem in strongly correlated electron systems, in particular, is at the forefront of condensed matter physics (e.g., high temperature superconductivity) and will remain a challenging problem well into the 21^{st} century. Pulsed neutron spectrometers are ideal for this purpose, as they can measure absolute intensities over wide ranges of Q and E. This is more easily achieved if SNS spectrometers can achieve superior sensitivity through the use of curved guides and/or guide halls.

In regard to the list of instruments discussed, the varied demands posed by the scientific problems require a correspondingly diverse instrument suite. The chopper spectrometers A.2 and B.1 would service much of the demand for lattice dynamics and momentum distribution problems, and address some of the coarser resolution experiments in fluids and amorphous materials. Problems that demand better quasielastic energy resolution, such as diffusional and relaxational phenomena, lead to the spectrometers A.1 or A.3. Interest in some other instrument types was shown for certain specific problems: a triple-axis type instrument B.3 (phonon dispersions), C.1 (relaxation in glasses), and B.4 (momentum distributions).

IIC. Chemical Spectroscopy

Participants:

Ken Herwig, ORNL/SNS
Bruce Hudson, Syracuse University
John Larese, BNL
Feri Mezei, LANL/HMI
Roger Pynn, LANL
Frans Trouw, LANL (chair)

Discussion:

This group identified three broad areas of interest: vibrational spectroscopy, diffusion in simple and complex systems, and Brillouin scattering. These three areas represent a large variety of chemical problems for which neutron spectroscopy represents a useful tool. A variety of topical scientific areas of interest to the chemistry community were outlined in the report on chemical applications of neutron scattering arising from the SNS meeting in Knoxville November 1998.

Some examples drawn from that report and appropriate here as well are:

- Vibrational spectroscopy of organic molecules, including radicals and large macromolecular systems (e.g., pharmacological and opto-electronic properties)
- Validation of quantum chemical computations by a direct comparison with neutron vibrational spectra (straightforward Q-dependence very useful)
- Vibrational spectroscopy of adsorbates at interfaces, related to catalysis and biological systems
- Diffusion of small molecules under a wide variety of conditions, particularly in systems with complex interfaces (e.g., clays, surfaces of hydrophobic and hydrophilic macromolecules, catalysts)
- Elucidation of the bonding in novel compounds, such as the dihydrogen complexes in inorganic coordination compounds
- Solvent structure and dynamics in restricted environments (mainly diffusion)
- Lubrication, selective adsorption, and diffusion at interfaces
- Local and chain dynamics of polymeric materials (vibrational and diffusion); validation of computational modeling approaches
- Brillouin scattering to characterize the hydrodynamic modes in fluids and macromolecular materials

The current and anticipated needs of the chemistry community lead to the following desirable characteristics for the SNS inelastic scattering instruments.

For vibrational spectroscopy, the next generation instruments should be designed for a better energy resolution and the ability to measure smaller samples. The synthesis of novel materials frequently is restricted to small quantities, and the current requirement for samples of hydrogenous materials on the order of fractions of a gram or more is too demanding. It is also the case that a broad range of energy transfer must be measured simultaneously with acceptable resolution across the entire range. This naturally leads to an inverse geometry spectrometer (B.2) which is also well suited to measuring small samples.

Most of the quasielastic scattering carried out to date has focused on hydrogenous compounds, but there are many problems involving diffusion in, for instance, ceramic materials such as fast-oxide conductors which are being considered in methane conversion plants. The success of quantum chemical modeling is well developed for the case of vibrational motion at low temperature in solids, and the next challenge will be to accurately describe transport properties. The Q-dependence of the quasielastic neutron scattering in novel and complex inorganic materials will be one of the critical tests of the next generation of quantum chemical models. Furthermore, the long-range diffusion issues in composite materials operated close to their design limits will require measurements over large fractions of Q and energy space. For diffusion in simple and complex systems this leads to the need for a Q-range of 0.001-5 $Å^{-1}$, in conjunction with an energy transfer range of 0.1 neV to 20 meV, with the best possible energy resolution (better than 10% of energy transfer in the worst cases). The Q-resolution requirement is typically modest at about 10%, but coherent quasielastic scattering problems will need much better Q-resolution in addition to good coverage in Q-space and a good energy resolution. These requirements cannot be satisfied by a single spectrometer, but a combination of A.1, A.3, and C.1 are essential for the future needs of the community, in the area of transport properties.

Current developments in chemistry and biology are moving towards systems of greater complexity, which implies greater length scales and excitations at lower frequencies. Brillouin scattering is appropriate for these kind of investigations, and it is likely that this technique will provide useful information on the dynamics in macromolecular compounds, where a local description of the dynamics alone will not provide the necessary insight. Such a spectrometer requires low momentum transfers (0.005 $Å^{-1}$) in conjunction with modest energy transfers and a good energy resolution (0.1-10 meV energy transfer and an energy resolution of 0.2-0.5%). This is probably only achievable using a direct geometry instrument, such as spectrometer C.2 in Table I.

A recurring theme was the need for a wide variety of ancillary equipment, and the desire to use multiple probes on a sample. The ability to measure a neutron diffraction pattern at the same time as probing the molecular dynamics of the system under study is an established and desirable requirement. This is straightforward to achieve in white beam instruments such as A.1 and B.2, but this should also be achievable on direct geometry instruments, if automated removal of elements such as monochromating choppers is possible. As the samples are frequently hydrogenous, the neutron diffraction data is of limited utility in those cases, and alternative techniques for *in situ* characterization of the sample is desirable. In principle X-ray diffraction would be useful in this context, but this is likely to be difficult to achieve in practice. However, there are a large number of analytical techniques in common use in chemistry, such as optical spectroscopy, nuclear magnetic resonance, electron spin resonance, and gas chromatography/mass spectroscopy. For example, the ability to simultaneously measure diffusion using magnetic resonance techniques and quasielastic neutron scattering would provide for an unprecedented ability to measure diffusion over many orders of magnitude on the same sample.

The most logical way forward for the development of such multiple probe techniques is to develop the concept of a "sample environment module" which can be assembled with the desirable options and samples. This would be done outside of the instrument, leading to greater control and reproducibility, and more importantly avoid the considerable waste of beam time which occurs during sample "preparation" (e.g. cooling, heating, irradiation with lasers etc.). This is likely to be a major issue with some of the faster instruments envisaged for the SNS.

For all three types of scattering the abilities to go to pressures of 0.1 Gpa was considered useful, as was continuous and pulsed (> 10Hz) magnetic fields (i.e., fields as high as possible, but consistent with the need for an acceptable count-rate). Furnaces capable of providing sample temperatures for measurements up to 2000K are also desirable, in addition to the more conventional cryogenic equipment capable of going down to about 1K. For quasielastic scattering, a shear cell will provide the novel ability to measure transport properties as a function of shear.

Finally, the issue of small samples needs to be addressed using neutron optical elements to focus the beam onto small volumes. It is also desirable to provide a polarized neutron option, in order to discriminate between coherent and incoherent scattering from complex materials.

III. Summary and Recommendations

After the breakout sessions of the three working groups, a general discussion session was convened to discuss the overall needs for inelastic scattering instrumentation at the SNS. The various science-based criteria were evaluated in the light of the likely capabilities of different types of neutron spectrometer designs (e.g., direct geometry and inverse geometry instruments). This discussion led to consideration of the 10 instruments listed in Table I.

The SNS project currently has three ongoing or planned design studies for new spectroscopy instruments for "day-one" operation of the SNS. These are listed in Table I, section A. On the basis of the scientific requirements articulated by the working groups, these three instruments are desirable to address a wide variety of scientific problems. After further discussion it was decided that a total of seven instruments, those listed in Table I-A and I-B, should be studied as potential "day-one" instruments.

The three remaining instruments (Table I-C) are not being recommended for further study at this time. The spin-echo spectrometer is desirable for a wide variety of scientific applications but there is the prospect that a SNS-Julich/HMI collaboration will develop this instrument for pulsed sources. It may be possible to accommodate Brillouin scattering spectroscopy on one of the other instruments. The PRISMA design can be evaluated as a feature on some of the other instruments to address their scientific requirements.

Appendix A: Workshop Agenda

Inelastic Neutron Scattering Workshop
Argonne National Laboratory
November 1, 1999

Location: IPNS, Building 360

8:30 am - 8:45 am: Greetings and Introductory Remarks
Thom Mason, Kent Crawford (A-224)

8:45 am - 10:15 am: Breakout into Science Interest Groups I
(i) Magnetism (Steve Nagler) (A-224)
(ii) Chemical Excitations (Frans Trouw) (G-223)
(iii) Thermodynamics, Lattice Effects, Critical Phenomena
(Paul Sokol) (G-219)

10:15 am - 10:30 am: Break (C-232)

10:30 am -12:00 Noon: Breakout into Science Interest Groups II

12:00 Noon - 1:00 pm: Lunch (Food in C-232, Dining A-224)

1:00 pm - 2:00 pm: Recommendations from Groups for
Instrumental Capabilities (A-224)

2:00 pm - 3:30 pm: Outline of Instruments to realize the Capabilities

3:30 pm - 3:45 pm : Break

3:45 pm - 4:30 pm : Compilation of priorities

Appendix B: Participant list

Name	Institution	Session
Doug Abernathy	ORNL/ANL	L
Ken Andersen	ISIS	L
John Ankner	ORNL/ANL	M
Ward Beyermann	U. of Cal. at Riverside	M
Collin Broholm	Johns Hopkins U.	M
Bruce Brown	ANL	–
Kent Crawford	ANL	–
Eugene Goreuychkin	ANL, JINR	M
Ken Herwig	ORNL/ANL	C
Bruce Hudson	Syracuse	C
Sandy Kern	Colorado St. U./ANL	M
John Larese	BNL	C
Chun Loong	ANL	M
Thom Mason	ORNL	M
Rob McQueeney	LANL	L
Feri Mezei	LANL	L,C
Steve Nagler	ORNL	M(chair)
Ray Osborn	ANL	M
Roger Pynn	LANL	M,C
Jim Rhyne	U. of Missouri	M
Sunil Sinha	ANL	M
Paul Sokol	Penn State U.	L(chair)
John Tranquada	BNL	M
Frans Trouw	LANL	C(chair)

M = Magnetism C=Chemical Excitations L=Lattice effects, thermodynamics, liquids…