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**"SCIENTIFIC OPPORTUNITIES WITH ADVANCED FACILITIES
FOR NEUTRON SCATTERING"**

Report of a Workshop Held at
Shelter Island, New York
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PREFACE

The Shelter Island Workshop brought together ~ 60 participants from a variety of countries, institutions, and scientific backgrounds to consider the needs for new more intense neutron sources, improved instrumentation at existing facilities, and detailed comparisons between instrument performance at steady state and pulsed sources.

The contributions from the individual groups (Chapter V) constitute the bulk of this report and the editors wish to thank the writers W. Koehler (ORNL), C. Windsor (AERE), J. Jorgensen (ANL), T. Koetzle (BNL), D. Price (ANL), R. Moon (ORNL), J. Hayter (ORNL), J. Eckert (LANL), R. Brugger (Univ. of Missouri), and S. Werner (Univ. of Missouri) for their conscientious effort in getting the manuscripts first written on time, and then circulating them to the other members of their group. The editors also wish to thank J. Axe for his constructive help.

The organizing committee for the Workshop was M. Rowe (NBS), R. Moon (ORNL), R. Silver (LANL), together with the two editors, and we wish to express our thanks to our colleagues for their help - materialistically for obtaining financial support from their home institutions and for advice.

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ABSTRACT

The present report documents deliberations of a large group of experts in neutron scattering and fundamental physics on the need for new neutron sources of greater intensity and more sophisticated instrumentation than those currently available. An additional aspect of the Workshop was a comparison between steady-state (reactor) and pulsed (spallation) sources. The main conclusions were:

- (1) The case for a new higher flux neutron source is extremely strong and such a facility will lead to qualitatively new advances in condensed matter science and fundamental physics.
- (2) To a large extent the future needs of the scientific community could be met with either a $5 \times 10^{15} \text{ n cm}^{-2}\text{s}^{-1}$ steady state source or a $10^{17} \text{ n cm}^{-2}\text{s}^{-1}$ peak flux spallation source.
- (3) The findings of this Workshop are consistent with the recommendations of the Major Materials Facilities Committee.

An abbreviated report of this Workshop is being prepared for publication in Nuclear Instruments and Methods in late 1985.

I. HISTORICAL BACKGROUND AND OBJECTIVES OF WORKSHOP

In late 1982 a Committee was formed under the auspices of the National Academy of Sciences to examine the "Current Status of Neutron-Scattering Research and Facilities in the United States". This Committee, under the chairmanship of Jack Rush (National Bureau of Standards) produced its report in late 1983 and it was published¹ in June 1984. Briefly, the report documented the disparity between neutron facilities in Europe and the U.S., discussed the need for new instruments and sources, and recommended a number of funding actions to increase investment. During 1983 the individual Laboratories with neutron sources made proposals for the upgrading of their own facilities, or the start of construction for new sources. These led to further discussions among the various agencies and the setting up by the Office of Science and Technology Policy of the Committee on "Major Materials Facilities", chaired by F. Seitz and D. Eastman. This Committee met in early 1984 and issued its report² in September 1984.

In these deliberations, the need for a detailed comparison between pulsed and steady state sources for various areas of scientific investigation became increasingly clear. This issue had been partly addressed in two small workshops, one sponsored by Argonne at Lake Geneva³ in February 1984 and the other by Oak Ridge in conjunction with their Users Meeting⁴ in May 1984. However, the idea for Shelter Island was to involve scientists from all the National Laboratories with major neutron sources and address these questions in a wider forum, also involving a variety of neutron experts from outside the U.S. The Shelter Island Workshop was thus sponsored by Argonne National Laboratory, Brookhaven National Laboratory, Los Alamos National Laboratory, the National Bureau of Standards, and Oak Ridge National Laboratory.

Organizing Committee

V. Emery, BNL, Co-Chairman
G. H. Lander, ANL, Co-Chairman
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References

1. Report on "Current Status of Neutron-Scattering Research and Facilities in the United States", published by National Academy Press, Washington, D.C. 1984.
2. Report on "Major Facilities for Materials Research and Related Disciplines", published by National Academy Press, Washington, D.C. 1984.
3. Proceedings of Workshop at Lake Geneva, Wisconsin, on "Instrumentation and Scientific Opportunities at a 10^{17} Peak Flux Pulsed Neutron Source", ed. R. K. Crawford, Argonne National Laboratory, (unpublished, 1984).
4. Report on "The Scientific Case for A New Higher Flux Neutron Source", ed. by R. M. Moon, Oak Ridge National Laboratory, (unpublished, 1984).

II. WORKSHOP PROGRAM

The Workshop opened with four talks on sources and instruments. The first by Jack Carpenter (ANL) described the parameters of the ASPUN spallation source that is expected to attain a peak neutron flux of $\sim 10^{17}$ n cm⁻²s⁻¹ with about 4 mA time-averaged proton current and a proton pulse length less than 1 μ s. Details of the neutron spectrum from a number of different moderators from this source are presented in Appendix 1 of this report. In the following two talks by Gunter Bauer and Hans Stiller, both from Jülich, the German SNQ source and some of the instruments designed for it were described. The SNQ source as now envisaged is based on a linac machine delivering ~ 4 mA time-average proton current at a repetition rate of 100 Hz and with proton pulses of 250 μ s length. The time-averaged flux of SNQ is 7×10^{14} n cm⁻²s⁻¹, with peak fluxes some 40 times greater than this. Hans Stiller discussed the instruments they have considered and how the pulse structure can be used to obtain "gain factors", as compared to instruments at a steady state source of comparable average flux, varying from 1 for a triple-axis spectrometer to ~ 50 for some machines using time-of-flight and cold neutrons. The average gain factor was greater than five.

The last talk in the Introductory Session was by Ralph Moon (ORNL) on the plans for a new reactor at ORNL. The goal of a reactor with a flux of 5×10^{15} n cm⁻²s⁻¹ appears feasible with current technology. According to present ideas, higher fluxes would require surface modification of the fuel. A description of the flux distributions emanating from this reactor is given in Appendix 2. Both Appendices 1 and 2 were available to all the participants before the Workshop and acted as resource documents.

After these introductory talks the participants broke into groups and confronted the various issues discussed above. All groups gathered for a plenary session half way through the Workshop and another summary session at the end. The group reports represent the bulk of this Proceedings and the participants to each one are listed on the first page of each group report.

Chapters III, (Neutrons in Condensed Matter), IV (The Need for More Intense Sources) and V (Summary and Recommendations) were produced by the Workshop Liaison Group, whose duties were to summarize activities and act as liaison between the other groups. The members of this group were J. D. Axe (BNL), V. Emery (BNL), B. E. F. Fender (ILL), G. H. Lander (ANL), S. K. Sinha (Exxon) and H. Stiller (Jülich).

III. APPLICATIONS OF NEUTRON SCATTERING

A. Neutrons in Condensed Matter Science

The power and utility of neutron scattering for the study of condensed matter has been well-established over the past 35 years. Neutrons are distinguished from other probes by a number of specific properties:

- (i) The energy (E) and momentum (Q) of incident neutrons may be matched simultaneously to the typical energies and length scales of processes occurring in condensed matter. The most convenient neutron wavelengths (1 -10 Å) are ideal for the study of interatomic spatial correlations, and available neutron energies (1 to 1000 meV) allow the measurement of excitations in the μ eV to meV range. (By contrast, 1 Å wavelength x-rays or electrons have energies ~ 12 keV and ~ 3.5 eV respectively).
- (ii) The neutron has a magnetic moment which couples to spatial variations of the magnetization density of materials on an atomic scale. Neutrons are thus ideally suited for the study of magnetic structures and short wavelength magnetic fluctuations such as spin waves.
- (iii) Neutrons interact with atoms principally through short ranged strong nuclear interactions, which vary erratically from nucleus to nucleus. Thus light atoms may be studied in the presence of heavy ones. Perhaps the most important single isotopic difference is between hydrogen and deuterium, because of its significance for biology and polymer science.
- (iv) In virtue of their low absorption by most materials, neutrons are a bulk probe.

In many applications, it is extremely effective to combine neutron scattering studies with x-ray, electron, or light scattering experiments or the developing technique of muon spin relaxation. The future opportunities for this approach have been dramatically affected by the availability of synchrotron x-ray sources, which have brought new possibilities for the enhancement and control of the major characteristics of a photon beam: source size, spatial and temporal coherence, spectral range, polarization and time structure. But neutrons and x-rays are truly complementary probes, and these developments provide an additional impetus for advances in neutron scattering. In order to exploit the capabilities and enormous potential of a broadly-based scattering program for condensed matter sciences, it is necessary to make provision for new sources of both x-ray and neutrons, as well as the instrumentation to make full use of them. The range of neutron scattering is represented schematically in Fig. 1 where we see that the technique extends over at least six orders of magnitude in both momentum and energy transfer.

B. Industrial Applications of Neutron Scattering

The use of neutron scattering research on condensed matter for industrial applications is still in its infancy, and has been historically hampered by both a lack of expertise in this area in most industrial research laboratories and lack of access to the major neutron scattering facilities in this country. However, this situation is rapidly changing as scientists in industrial laboratories become more familiar with the scope of neutron scattering research, and as the National Laboratories institute procedures for greater access to the neutron scattering facilities. We thus may anticipate a large increase in neutron scattering research applied in industry over a wide range of problems ranging from basic to applied research. Because of the importance of hydrocarbons and H-containing materials in a large number of industrial processes, it is becoming increasingly recognized that neutrons have a unique role to play by providing an advanced probe for the study of the complex materials and processes on which future advanced technologies will be based. A new intense neutron source could open up vast new areas of research on these systems. For instance, from the industrial perspective, there is much interest in the structure and conformation (including wetting properties) of polymers or hydrocarbon liquids and gases on surfaces, intercalates, clays, in disordered systems such as porous media and in bulk solids on length scales from 10 Å-5000 Å, which falls awkwardly between the regimes accessible to conventional small angle and light scattering experiments.

Fig. 1. Schematic representation of some of the fields of science covered by neutron scattering. The fields are roughly represented by the area in which the words appear. The center-shaded-area represents elastic scattering with no energy discrimination.

The same is true for the growth of precipitates, voids and cracks in wet samples (particularly in real time) under the influence of stress or quenching. The H/D contrast variation property of neutron scattering and the penetrating power of neutrons for metallic samples makes this particularly attractive on the new generation of neutron sources rather than at the new X-ray sources.

Also of great interest is an understanding of the microscopic processes whereby molecules diffuse in porous media, membranes, or generally in systems with restricted geometry. The use of high energy and momentum resolution inelastic neutron scattering coupled with the ability to do experiments at small Q values would lead to a breakthrough in our understanding of the processes relevant to transport, phase separation, and catalytic activity in a wide variety of complex technologically important materials.

C. Fundamental Physics

In another vein, the study of the wave and particle properties of the neutron is of great interest for fundamental physics and has a long tradition. Experiments of current interest would give a better determination of the neutron lifetime, continue the search for a neutron electric dipole moment and for neutron-antineutron oscillations and improve our knowledge of the neutron-neutron interaction. These and a variety of interferometry experiments have an impact on nuclear physics, particle physics, astrophysics and cosmology.

IV. THE NEED FOR MORE INTENSE NEUTRON SOURCES

Neutron scattering is a field of enormous breadth, ranging from applications in molecular biology to fundamental physics. This range is demonstrated by the Figure in chapter III above, where the units show that neutron scattering covers more than 6 orders or magnitude in both energy and momentum transfer. In a field such as this, no single experiment, or even a series of experiments, can be identified as the most important in condensed matter. The present Proceedings show emphatically that, with new sources of the type proposed, completely new experiments will be possible on materials of central importance in condensed matter science. The individual sections discuss many of these examples, some in considerable detail. In this section we discuss the general areas into which the new experiments will be possible.

(a) Resolution

Increases in resolution with new sources and instruments do not simply allow present experiments to be performed more easily; they open up completely new

areas. For example, although spin-echo and back-scattering instruments have permitted major advances in areas such as polymer dynamics and tunneling spectroscopy by achieving μeV resolution close to $E = 0$, these techniques are not as yet suitable for studying dispersive excitations. For dispersive modes $\Delta E/E \sim 0.01$ is considered tolerable because of the compromise required between intensity and resolution. This resolution is poor by standards of optical techniques that routinely achieve $\Delta E/E \sim 10^{-4}$. The lineshapes of collective excitations as measured by neutron scattering contain important information but are presently instrumentally limited. One important example is the electron-phonon interaction. For phonons with energies below the superconducting gap, 2Δ , the electron-phonon interaction vanishes in the superconducting state because these phonons are not energetic enough to break Cooper pairs. The changes observed in the line widths of these phonons through T_c provides a direct measure of the electron-phonon interaction. To date such measurements have been attempted only for specially selected "demonstration" cases. Increased resolution will vastly expand the scope of the technique.

Improved energy resolution is of importance in studying single particle excitations as well as collective modes. Epithermal neutrons at sufficiently high momentum transfer produce individual particle recoils (atomic Compton scattering). The mean position of these recoil peaks is determined by scattering kinematics but the intrinsic shape of the peaks measures the momentum distribution, $n(p)$, of the particles in their initial state. The determination of $n(p)$ is of great interest for the quantum fluids (He^3 and He^4), and also for very anharmonic solids (e.g. H-bonding), but present resolution is sufficient to reveal only the most general features of the distributions.

Exceptionally high energy resolution near $E = 0$ is presently achieved by spin-echo and back scattering, but at the expense of relatively poor Q resolution. The importance of energy resolution for small- Q scattering machines and others that normally aim simply for good Q space resolution is beginning to be appreciated. These machines can be instrumented to perform inelastic small- Q scattering which will make possible completely new experiments in liquids, amorphous, and magnetic materials.

The minimum Q presently attainable with neutron small-angle scattering is $\sim 10^{-3} \text{ \AA}^{-1}$, which is coincidentally about the maximum attainable with light scattering. Thus the extension of neutron techniques to $\sim 10^{-4} \text{ \AA}^{-1}$ will allow good overlap between these complementary techniques and open up completely new ranges of investigation with neutrons. Increased Q resolution is of central

importance in biological and chemical investigations. In SANS experiments extrapolation to $Q \rightarrow 0$ for molecular weights $M_w > 10^6$ and Guinier radii of $\sim 10^3$ Å require minimum Q values of $\sim 10^{-4}$. A good example is of polymers of the type $(PVF_2)_n$, where those of technical importance have $MW > 10^6$. Both momentum and spatial resolution is of importance in many areas in applied materials science such as residual stress and texture. Neutrons represent a potentially powerful technique in this area because of their penetration.

Temporal resolution will be of importance for studying transient states and can only be obtained at the expense of intensity. With existing sources the intensities are too low for any but the simplest of real time experiments, but we see this as a rapidly growing area. The attainment of much higher intensity SANS instruments will also permit the kinetics of processes such as precipitation, phase decomposition, and grain coarsening to be followed in real time, on a scale of 0.1 sec or greater, under realistic conditions of temperature and stress. Further examples are the real time relaxation of macromolecules and transients induced by external stimuli such as laser induced photochemistry.

(b) Small Cross Sections and Differential Scattering

In almost all applications neutron scattering is an intensity limited technique. This means that even for experiments performed at present sources the experiments would benefit directly from more flux. However, past experience has shown that increases in intensity open new scientific areas, and we anticipate similar opportunities with the next generation source. The neutron cross section is small (this fact frequently makes for an unambiguous interpretation of the results) so that rather large samples have to be used. Many materials of interest are available in only very small quantities, or the scattering centers of interest are dispersed dilutely. Examples of new materials in which neutrons could play a unique role with more intensity are: the observation of spin density waves in organic conductors; diffusion studies with dilute systems such as hydrocarbons in zeolites; diffusion studies involving atoms other than hydrogen; conformation studies on dilute biological systems such as nucleosomes and viruses in which low concentrations are necessary to approach those of physiological interest; the dynamical behavior of monolayers and intercalates and artificial multilayer structures; and studies of both the structural and dynamical behavior of materials at high pressure (above 50 kbar). Many of these are discussed in detail in the sections that follow. One of the biggest uncertainties lies in the area of biology, where sample size usually represents the most important limiting feature. Coupled with the necessity to use deuterium

labeling we can easily predict that the availability of neutron fluxes of sufficient intensity to use the same samples as in x-ray, light scattering, or NMR experiments, will lead to a qualitative leap in our understanding of these materials.

The term "differential scattering" can be applied to a wide variety of experiments done today with neutron scattering in which small changes between two data sets need to be measured accurately. For example, to measure the effects of vacancies in crystal lattices one needs a series of experiments at different vacancy concentrations. More typically, the sophisticated experiments make use of isotopic substitution to change the scattering potential without changing the chemistry. Increases in intensity will allow the isotopic substitution of atoms such as C and N for which the differences in scattering lengths of isotopes of the same atom are very small. For biological and polymer systems the common substitution is deuterium for hydrogen. Although the difference in the scattering lengths of these isotopes is large, there is a need for selective deuteration at very low concentration. Investigations involving inelastic scattering of biological systems also involve differential scattering and selective deuteration. In all these applications higher fluxes are needed to give the required statistical accuracy.

(c) Polarization Analysis (PA)

Polarized neutron techniques allow for the separation of magnetic and nuclear scattering as well as coherent from incoherent scattering arising from nuclear spins. Examples of studies that would benefit from PA are excitations in magnets above their ordering temperature, the spin response functions of intermediate valence and actinide materials including the new heavy fermion systems, and the dynamics of spin glasses, amorphous magnetic materials, and investigations of magnetic defects in antiferromagnets. These examples utilize the ability to separate the magnetic from the nuclear scattering - equally important will be the ability to separate coherent and incoherent and thus learn about the self and collective terms in diffusion processes in a variety of materials, the most important of which will almost certainly be in the fields of chemistry and biology. The full exploitation of the neutron interaction requires that the neutron spin vector be determined both before and after the scattering process. In practice even polarization analysis (PA) of one spin component is usually sufficient to obtain a great deal of information about the scattering process.

The promise of PA has, of course, been present for many years. Unfortunately providing polarized beams and analyzers is costly in terms of neutron intensity and difficult technically for neutrons shorter than $\sim 2 \text{ \AA}$. However, we identify PA as a "threshold" field that will rapidly expand with the next generation source. There are good reasons for this optimism; over the last decade an increasing number of instruments have been built to incorporate PA, and an increasing effort is being expended into developing polarization devices for both cold and hot neutrons. We note also, for example, that the first use of PA to separate the incoherent and coherent inelastic scattering in a liquid (molten Na) was recently performed at the ILL. Thus, the next generation source will open up entirely new areas, as well as add clarity to many existing ones, by having PA as an option on at least 50% of the neutron instruments. The present level (at ILL for example) is $\sim 5\%$, but increasing to $\sim 20\%$ on the new instruments being designed.

V. SUMMARY AND RECOMMENDATIONS

Three important points emerged from the Shelter Island Workshop:

- (1) **The case for a new higher flux neutron source is extremely strong and such a facility will lead to qualitatively new advances in condensed matter science and fundamental physics.**

The general areas of scientific progress are highlighted in Chapter IV, and discussed more extensively in the reports of the individual groups, Chapter V.

- (2) **To a large extent the future needs of the scientific community could be met with either a $5 \times 10^{15} \text{ n cm}^{-2}\text{s}^{-1}$ steady state source or a $10^{17} \text{ n cm}^{-2}\text{s}^{-1}$ peak flux spallation source.**

However complete overlap in the capabilities of the two sources does not exist, and in some areas the sources are truly complementary.

Important areas in which the steady state source is superior are (a) studies of collective excitations, which often require moderately high Q - and E -resolution simultaneously, and for which triple axis spectroscopy is well suited; (b) the extension of SANS to very small momentum ($Q < 10^{-4} \text{ \AA}^{-1}$). Conversely, the pulsed sources will be superior for high-energy and high- Q scattering, both elastic and inelastic, and for certain real time studies. The weight assigned to such individual areas of science is to a certain degree subjective and this Workshop made no effort to make such judgements.

- (3) **The findings of this Workshop are consistent with the recommendations of the Major Materials Facilities Committee.**

On the basis of their considerations of the scientific opportunities, the Major Materials Facilities Committee of the National Research Council recognized the importance of continuing development of both steady state and pulsed sources. They recommended design studies for a $5 \times 10^{15} - 10^{16} \text{ n cm}^{-2} \text{ s}^{-1}$ advanced reactor to begin immediately, leading to construction starting in FY'89. Concerning the construction of an advanced pulsed neutron source the Committee recommended that the decision follow further assessment of the performance of existing and newly operating lower-flux pulsed sources. However, this Workshop documents the rapid pace of development, especially associated with the German SNQ project, of innovative instruments designed

for research with cold neutrons at advanced pulsed sources. Given sufficient time for construction and thorough evaluation, many of these instruments will undoubtedly greatly extend the already substantial capabilities of pulsed sources.

In designing instruments for a new facility the group became aware of a number of technical limitations that prevent further progress in instrumentation. In order of priority these are:

- (a) The need to develop supermirrors and multilayers for the transportation of thermal ($1 < \lambda < 3 \text{ \AA}$) neutrons.
- (b) The need to develop high speed position sensitive detectors. The best such detectors now have dead times of $\sim 400 \text{ ns}$, but, especially for an advanced spallation source, detectors at the level of $\sim 100 \text{ ns}$ will be needed to handle the peak count rates envisaged.
- (c) More effort needs to be made on polarizing techniques, especially for white beam applications. The general field of polarization analysis will not become available, especially at pulsed sources, until further advances are made in this area. We should stress here that this refers to wavelengths less than $2 \text{ \AA}$, for $3 \text{ \AA}$ and longer wavelength neutrons devices currently available are satisfactory.

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Small-Angle Scattering

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Scientific justification for small-angle neutron scattering (SANS) instrumentation on new sources, both pulsed and steady state, has appeared recently in committee reports,^{1,2} workshop proceedings,^{3,4} and the scientific literature.^{5,6} The most recent is the report entitled "The Scientific Case for a New Higher Flux Neutron Source, Part I-Neutron Scattering" prepared at Oak Ridge in October, 1984 and distributed to all the Shelter Island participants. Scientific disciplines that regularly use SANS include metallurgy, polymer science, physical chemistry, glasses and other materials sciences, solid state physics, and biology. Significant new science is continuously being identified in these and other research areas.

One difficulty in planning new SANS facilities is that the terms SANS and SANS instrument mean different things to people working in different fields of research. No one instrument can satisfy the needs of all types of experiments, and justification can be made for several types of instruments on either an advanced reactor, a pulsed source, or both. In the proceedings of the ANL workshop³ two different pulsed source SANS instruments were recommended. Need for four different reactor based SANS facilities was identified at the ORNL workshop.⁴ These several instruments have different requirements for Q-range, minimum Q attainable, resolution in Q, and capability for polarization and/or energy analysis. Another important consideration in instrument design is that of sample size. This ranges from several square centimeters in some SANS research areas to less than a square millimeter in others. One extreme case is the use of SANS for low resolution crystallography of biological macromolecules (such as viruses and membrane proteins) where the sample volume may be only 0.1 mm^3 . Requirements for collimation distances, beam sizes, detector and pixel dimensions, and sample-to-detector distances are very different over this large range of sample dimensions.

In spite of this wide range of experimental requirements several general characteristics of new SANS facilities can be identified.

1. New high flux sources must provide copious quantities of useful cold neutrons. Most experiments are carried out at wavelengths ranging from 4–12 Å. Frequently even longer wavelengths, up to 20 Å, are desirable. At present, reactors with suitable cold sources are the best sources of neutrons in the wavelength range 3–20 Å. Spectra from cold moderators on pulsed sources also contain neutrons in the wavelength range from three to ten Å, and instruments are in use, (IPNS or KENS) or are being designed (WNR/PSR, SNS, SNQ) on such sources. However, SANS measurements on pulsed sources have usually used shorter mean wavelengths than the equivalent experiments on reactor based instruments. Detailed intensity comparisons of machines on steady-state reactors and pulsed sources depend upon the particular types of experiments being performed and they have been difficult to make. Extremely important for the intercomparison of instruments is the Q resolution of the measurement.^{7,8} In a balanced reactor based instrument the counting rate at the detector varies as the fourth power of the resolution ΔQ , fifth if the contribution of $\Delta\lambda/\lambda$, which is important at large Q , is included. On the pulsed source instrument, the geometrical contributions to ΔQ are similar to those of the reactor based machine.

To obtain a given scattering vector, the pulsed source instrument must on the average use scattering angles that are smaller than those required for longer wavelength reactor based instruments and much more stringent collimation is required. In particular the attainment of very low Q values, $\sim 3 \times 10^{-4} \text{ Å}^{-1}$, is best done on the reactor based instrument. For a reactor based instrument $\Delta\lambda/\lambda$ has to be appreciable, 6% to 20% is not uncommon, in order to obtain intensity. As a result ΔQ increases with Q , especially at the larger values of Q that are required for some experiments. On a pulsed source $\Delta\lambda/\lambda$ is negligible and the resolution ΔQ is independent of the scattering vector.⁹ The pulsed source is therefore advantageous in this regard in the higher Q part of the scattering pattern.

2. The time structure of the pulsed source is conveniently used to broaden the Q range measured in a SANS experiment because a range of wavelengths is used simultaneously. This can be particularly advantageous in some types of

experiments. Wide Q ranges can be covered in today's reactor based machines, but at the cost of doing multiple experiments (change the wavelength, or sample-to-detector distance) and combining the resulting data, or perhaps moving to a different instrument entirely. These procedures are not satisfactory if short times to complete the measurements are required.

Alternatively, it is conceptually possible to incorporate innovative use of multiple detectors in a new SANS instrument on an advanced high flux reactor to span wider Q ranges simultaneously but, as stated above, such an instrument would have ΔQ increasing with Q in the high Q region.

A potentially useful application of the wide wavelength range of pulsed source instruments is that of measuring resonant scattering from systems suitably tagged (with Gd, for example). Scattering both above and below the resonance can be done in a manner analogous to that of anomalous scattering with synchrotron x-rays. The technique has possible general application since chelating agents with high affinities for resonant scatterers can be linked chemically to many macromolecules.

3. Important applications of SANS that are developing and will certainly continue are the study of time dependent phenomena and the study of systems in non-equilibrium states. Much of the work going on in these areas now is being performed at reactors but pulsed sources, with their time structure, may have applications to phenomena with time constants of a few milliseconds or less. Such short time constants are difficult or impossible to measure on present day reactor based SANS instruments for experiments requiring $Q_{\min} \sim 3 \times 10^{-3} \text{ \AA}^{-1}$. The sample-to-detector distances and $\Delta\lambda/\lambda$ requirements are such that the spreads of neutron flight times are comparable to the time constants.

4. In all of the documents extant that call for advanced neutron sources there is the recognition of the need for polarized beams of high intensity and some form of polarization analysis. The production of monochromatic beams of wavelengths useful for SANS at a reactor is relatively easy and polarization analysis in one-dimension seems to be quite possible, as for example, at the IN11 instrument at the ILL. Polarization analysis with a 2-D detector may also be possible with clever application of optical devices. In any case, polarized neutron SANS is not likely to be used routinely and will be an option to be

exercised as the experiment requires. Development of methods for producing polarized white beams, at the pulsed sources, is being actively pursued. If a simple filter can be found, it may also be used as an analyzer but SANS with polarization analysis on the pulsed source machines seems remote.

5. Energy analysis of the scattered beam in SANS facilities is probably more useful for pulsed-source instruments than for reactor-based ones because of inelastic processes possible at short wavelengths. It can be accomplished by phasing a chopper to the pulsing of the source. Similarly, in a new instrument at an advanced steady state source, coarse choppers can be used to provide energy analysis. For most purposes this option would probably not be used routinely.

II. SUMMARY

On the whole, most of the SANS experiments that are being done, and those that are extrapolations of our present experience, are best done at a suitably designed and equipped reactor. It is important to keep in mind that instrumentation development for intense pulsed sources is in an early stage but there are already demonstrated applications where the large Q range and the time structure are extremely important.

It is probable that different experiments, exploiting the special characteristics of the sources, will be done at the two types of facilities. Undoubtedly both will benefit from the ingenuity and creativity of the scientists anxious to use them.

III. SCIENTIFIC OPPORTUNITIES FOR SANS

SANS has proved, since its beginnings, about twenty years ago, to be applicable to an extremely wide range of problems. Use of SANS has risen explosively and wherever instruments have been built time on them has quickly become oversubscribed. Indeed, in many cases progress in the science done with SANS has been limited as much by the lack of accessibility to instruments on existing sources as to the development of new higher flux sources and new

types of instrumentation. After a decade of intense activity one can now begin to identify areas of investigation that really require the higher fluxes that new sources can provide. Accessibility will probably always be a problem and care must be taken to provide for a sufficiently large number of instruments.

In the following are listed several areas that will benefit from the intense sources under consideration.

1. Low-Q.

There is a general consensus that the capability of making SANS measurements routinely at lower values of Q than is done at present is desirable. In the conventional instruments that are currently in operation in the United States $Q_{\min} \approx 3 \times 10^{-3} \text{ \AA}^{-1}$ seems to be a realistic value at which clean data are obtainable (D 11 at Grenoble is capable of $Q \sim 3 \times 10^{-4} \text{ \AA}^{-1}$ but it is not often used in that configuration). In specialized applications a double-perfect-crystal-diffractometer may be able to go to $5 \times 10^{-4} \text{ \AA}^{-1}$ with a resolution $\Delta Q \sim 4 \times 10^{-5} \text{ \AA}^{-1}$. With another decade reduction to $Q_{\min} \sim 3 \times 10^{-4}$, some important problems could be investigated. A partial list follows:

- i) One needs small Q in order to make accurate extrapolations to $Q = 0$ to measure size distributions of polydispersed particles. Such extrapolations are needed to characterize, for example, growth stages of creep cavities in metals and ceramics and of precipitates in metal alloys. They are important as well in the study of micellar and microemulsion systems. Often these size distributions are too broad to be characterized on current instruments.
- ii) One needs lower Q values with high resolution to measure long (longer than possible at present) range periodic structures. These may include magnetic superconductors, fluxoid lattices in superconductors, or lamellar structures in phase separated polymers. Many magnetic systems exhibit spin density wave configurations which are only accessible at small Q .
- iii) A large Q -range, particularly pushed to small Q , is needed to characterize materials that have a self-similar structure over orders of magnitude in scale (fractals). These include the ramified structures one finds in aggregates and porous materials with irregular interfaces.

iv) It is frequently desirable to measure polymer molecules or biological entities with molecular weights ranging from 10^4 – 10^7 with characteristic radii of gyration lying between 10–1000 Å. In most cases data obtained in the range $0.3 \leq QR_g \leq 0.5$ Å are desirable. The larger value of Q is no problem but with $Q_{\min}$ restricted to 4×10^{-3} Å⁻¹ one is very much restricted in the size (molecular weight) of objects one can study. An example is (PVF₂)_n for which applications of technical importance exist only for molecular weights in excess of 10^6 .

v) More and more, interest is being expressed in the response of systems to various kinds of external perturbations. In the case of stretched polymers, or aligned colloids or biopolymers, data are taken in, generally, two perpendicular directions. The data are evaluated in only a small part of the total area of the area detector, and the advantage gained by radial averaging is not available. These considerations are not of course, restricted to low Q .

2. Extended Q-range.

The importance of an extended Q range has already been mentioned. Some specific applications, without reference to how this extended range is to be achieved, are as follows:

i) In a fiber type nematic phase (liquid crystal type polymers) there is an ordering that produces diffuse Bragg peaks and there is intensity at small Q from the form factors and structure factors of the polymer molecules. The intensities are generally weak and the molecules are easily degraded. One needs to be able to record data as fast as possible over a Q range from 10^{-3} Å⁻¹ to several Å⁻¹.

ii) There is a great need to investigate the microstructure of materials. The properties of metals, alloys, ceramics, glasses in bulk, powder, or thin film applications are strongly dependent on the microstructure. This microstructure can in many cases be controlled if attention is paid to the very early stages of its formation, that is, to nucleation, spinodal decomposition or chemical precursors in the processing stage. In these early stages signals are weak and spread over a wide Q range. In some cases of

interest, oxide glasses for example, the study of the early stages requires a very wide dynamic range $Q_{\max}/Q_{\min} \geq 20$ because the data contain information on interparticle interference. Further, the scattering is very weak due to the small particle size.

iii) Scattering from oriented samples gives increased information over solution scattering. It is known that partial alignment of long molecules can be achieved with stress, shear, electric, or magnetic fields. The high flux and wide Q range required for spontaneously ordering systems, as in i) apply to these systems also.

3. Improved Resolution

In some experiments of current interest one would trade intensity for increased resolution. Included in this application are studies of the internal structure of complex, monodispersed particles (micelles, compound latex particles, viruses) and interface structure of phase separated block copolymers, for example. To investigate interactions between large assemblies in biological systems one requires simultaneously high first order resolution, high order to order resolution, and an extended Q -range.

4. Polarization Analysis

Polarization analysis will be important in many SANS studies where most of the background, frequently high, is due to incoherent scattering from protons in the sample itself. Polarization analysis permits direct, model-independent evaluation of the incoherent level. The technique has obvious application to magnetic systems, for instance, to studies of magnetic colloids (ferrofluids) and to the new class of systems formed by dispersing supramolecular particles in ferrofluids to obtain a variety of arrangements as a result of the effective magnetic interactions. The separation of magnetic and nuclear scattering by polarization analysis can be carried out in SANS just as well as in wide-angle scattering experiments. Up to now the general application of polarization analysis has been restricted, by low intensities, to high flux reactors.

5. Small, Dilute, and Thin Samples. Small Cross Sections.

The case for high fluxes to permit study of small samples is well documented in the area of biological studies. The quantities of materials available are nearly always tiny, due to the difficulty and expense of growing, separating, and purifying systems of interest. Sample areas in some newly designed instruments are of the order of 1 mm^2 . Other examples of hard-to-prepare or expensive-to-obtain samples are plentiful whenever one wishes to selectively deuterate specific molecular sites. One may align polymer molecules, as mentioned earlier, in flow cells. There is current interest in the study of polymer conformation in the aligned state but in a zero velocity gradient. This is produced in counter flow cells in which case the area of sample observed is of the order of 1 mm^2 .

There are numerous cases of interesting systems with low contrast that require high fluxes for their study. Samples that are at or near contrast match point with only a subgroup or absorbate scattering are examples.

A new field of investigation is that of the physical properties of thin films deposited on surfaces. One such problem is the low-angle reflection produced by a polymer film in semi-dilute solution. It is in principle possible to determine the density profile of such a film by total reflection using labeling of some particular layer in the film. This technique may have application to composite materials. It requires precise collimation (a few milliradians or less) and small samples (one or two cm^2 of surface). High flux at long wavelengths is necessary.

The scattering law for polyelectrolytes goes to a very low value at $Q = 0$. Up to now, because of intensity requirements for reasonable statistics, measurements have been made only in the semi-dilute range. To study the structure factor of the isolated noninteracting molecule a very low concentration is required (one in 10^4 or less).

Many experiments in which deuterium labeling plays a role are the type called differential scattering. The deuterium label is usually a small component whose location is to be determined. Consequently there are only small differences between the scattering from labeled and unlabeled samples. High statistical accuracy is required to clearly establish the location of the label.

6. Real-Time SANS

Use of SANS for real-time studies is a rapidly expanding one. One can measure the response of a system to a single-shot change in some external perturbation, or to the evolution of growth processes in various physical systems. In polymer blends and some metallic alloys spinodal decomposition occurs within seconds after quenching into the appropriate region of the phase diagram. The need for higher fluxes arises in those cases where the one-shot event to be time-sliced is very rapid, finishing in a second or so.

Another type of experiment uses periodic perturbations in which some change is applied many times to accumulate sufficient data. In polymer deformation experiments one wants to know how the polymer chain returns to its equilibrium configuration after stretching. Relaxation times range from milliseconds or less to several seconds. Kinetics of metal fatigue and void formation are similar experiments.

A particularly interesting application of real-time SANS is to the study of instantaneous local neighbor rearrangements in colloidal dispersions. Ion clouds around and ion condensation onto macroions might be investigated by contrast variation permitting a direct measurement of local potential in macroionic solutions. One can hope, with higher fluxes of cold neutrons to study processes in which hydrogen and deuterium are exchanged in biological molecules. It may also be possible to study hydrogen-deuterium exchange kinetics in single crystal proteins at low resolution.

Other investigations such as in-situ studies of the aging kinetics of precipitate growth and morphology in metal alloys, the kinetics of the sol-gel process and of the densification of ceramic powder compacts during sintering, and the response of ferrofluidic systems to applied magnetic fields are likely candidates for future experiments.

These examples of scientific opportunities are, almost necessarily, those that we see at short range today. It is inevitable that as new tools become available entirely different uses of them will appear. We can only try to provide the instruments and sources that will be flexible enough to respond to the needs of the future.

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Collective Excitations

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Summary

1. Collective excitations will remain the area from which new insights and concepts are obtained at the forefront of condensed matter physics.
2. Most experiments leading to major seminal developments will be challenging. They demand equipment that can focus on particular Q and ω , and provide the flexibility to change resolution, the focussing, and external parameters such as temperature, pressure and field. The flexibility that is essential is best provided by the triple axis spectrometer at the steady state source. It is recommended that all reactor instrument designs have fully optimized beam optics and polarization options.
3. A minority of experiments such as measurement of dispersion relations will require inelastic scans along symmetry lines in Q -space. Such experiments may well be suited to spectrometers with multiple analyzer arms. It seems likely they can equally well be deployed on reactor or pulsed sources. Although a comparable performance is anticipated, further study of the steady state and pulsed types of multianalyzer spectrometers is recommended, so as to permit a realistic assessment to be made. It is recommended that the feasibility of varying the incident resolution on a pulsed source, by variable poisoning, be investigated.
4. A minority of collective excitation experiments will require surveys of continuous distributions in Q, ω space. They are best performed on a rotor time-of-flight spectrometer on a pulsed source.
5. Benchmark experiments on a standard crystal e.g. Nb should be carried out at various laboratories with pulsed and steady state sources. Calculations to confirm the measured intensities will ensure that all factors affecting performance are understood.
6. Because of the crucial importance, high relevance and demand for experimental studies using incident energies less than 100 meV, it is recommended that, of the advanced sources we have considered, the reactor is the most advantageous source for the study of collective excitations.

1. Introduction

Perhaps the most important contribution of neutron scattering to the study of condensed matter has been through the study of collective excitations. Neutron scattering has provided crucial information about lattice vibrations and interatomic forces, magnetic excitations and magnetic interactions and the fluctuations occurring near phase transitions. These studies have been possible because the energies and wavelengths of thermal neutrons are ideally matched to the energies and wavelengths of the collective excitations in solids.

We are certain that thermal neutron scattering will continue to provide essential and unique information about the nature of the excitations and interactions between the constituents of materials. This is information which cannot be obtained by any other technique.

It is useful to classify experiments on collective excitations and phase transitions into three groups. These are those in which (type A) the experimenter is interested in studying in detail a very restricted and prespecified range of wavevector transfers Q and frequency transfers ω . Examples of this type of experiment are the study of critical scattering at phase transitions, and of the phason excitations in incommensurate materials. There is then a class of experiments (type B) in which information is required along a line in Q space, usually corresponding to a high symmetry direction and for a wide range of ω . A typical experiment might be the phonon dispersion relation in a new material. Finally the third class of experiments (type C) is that in which information is required over a large region of Q and ω . An example of this type of experiment might be the study of $S(Q, \omega)$ in superionic materials, paramagnets or a plastic crystal.

We are certain that in the future there will be a need to perform each type of experiment and that in each category there are experiments which require or will benefit from an increased flux. An example of an experiment of type A is gap spectroscopy in superconductors, seen either via the phonon or the magnetic excitation spectrum. Another is the study of charge and spin density waves in one-dimensional conductors for which only very small specimens are available and the study of critical scattering with a resolution sufficiently good that it will no longer be necessary to assume theoretical forms to analyze the data. Experiments of type B are the study of phonon and magnon dispersion curves in new materials which will provide basic information for materials science about interatomic and magnetic forces. An early experiment of this type is often a prerequisite of a more detailed later study of type A. Finally, experiments of

type C will enable detailed studies to be made of the dynamics of complex systems such as incommensurate systems, plastic crystals and of superionic materials.

It is anticipated that there will be the largest demand for experiments of type A. In the most recent ILL report roughly 2/3rds of the experiments on collective excitations fell into the type A category and 1/6th each into types B and C. It is our expectation that the most exciting and demanding experiments will be of type A. They will provide the seminal developments in the applications of neutrons in condensed matter. The concepts that emerge will have wide application in particle and non-linear physics.

Most studies of collective excitations are at present performed with triple axis crystal spectrometers at reactor sources. The advantages of this type of instrument for this work are discussed in detail in section 3.1. We consider that it will continue to be the most useful instrument for experiments of type A. Two novel spectrometers have been developed for pulsed sources to give information for fixed Q or along a line in Q space. These are the Constant- Q spectrometer and the MAX spectrometer. The use of these spectrometers for experiments of type A and B are discussed in section 3.2, where they are also compared with the triple axis crystal spectrometer with one detector and also with a multiple analyzer system but on a continuous source. Experiments of type C, which require information over a very wide range of Q and ω are usefully performed with chopper instruments which are described in section 3.3. They are compared in section 5.

It is envisaged that a new high flux source will operate largely in a user mode, probably along the lines of the ILL. The average length of an excitation experiment at ILL in 1983 was 8 days and it is unlikely that it will be longer on a new source. This means that, for difficult experiments which require the spectrometer to be well matched to the problem, it must be very easy to modify the experimental configuration, if several configurations are to be tried in the allotted experimental time. We therefore consider flexibility and the ease with which the resolution can be controlled to be very important.

2. Scientific Justification

The most exciting way to exploit the increased flux of the proposed new sources is by pushing the resolution in Q and ω into regimes which are not accessible with conventional instruments on existing sources. A selection of these problems follows:

- (i) Studies of gaps in superconductors. The effects of superconductivity gaps on phonon linewidths have been studied in Nb_3Sn and Nb , two relatively-high T_C ($T_C = 18$ and 9 K) materials. The gaps are about 5.5 and 3 meV. No measurements have been performed on other superconductors with low T_C 's. In light of the interest in the heavy fermion systems, where T_C 's are less than 1°K and gaps are less than 0.5 meV, both higher fluxes and better resolution are needed. Higher fluxes are needed because these effects occur at lower temperatures, with a corresponding decrease in response, and better resolution is needed because the energy is lower.
- (ii) There has been considerable interest in the 1-D organic compounds especially since some of them, for example TMTSeF-Br , have revealed spin density wave (SDW) transitions. However the SDW's, which should be observable by neutron spectroscopy, have yet to be observed. This is due to small sample size, low expected signals and the inability of theorists to define the positions in Q space where SDW's appear. With more intense sources the above restrictions may be removed and a more definitive limit set on the magnitude of the SDW.
- (iii) Only recently have spin waves been measured in Fe up to 160 meV. These measurements have been performed at the IPNS and other measurements in Ni have been performed at the hot source at ILL up to 200 meV. At these energy transfers, intensity becomes a limiting factor. There is strong interest to pursue these spin waves to higher energies and to establish where the spin wave spectrum merges with the Stoner continuum. This has been observed in some low T_C materials (eg MnSi) but not for the simple monatomic materials. Related measurements are the studies of paramagnetic scattering in these materials. The change in $S(Q, \omega)$ from the ordered state to the disordered paramagnetic state has been measured for small Q and ω and it would be desirable to push these towards the zone boundary. Also, the use of polarized beams to study these problems suffers from inherent inefficiencies of monochromators and analyzers and more flux is needed.
- (iv) Plastic crystals have been studied by ultrasonic methods and the velocities of sound measured. However, in the Q and ω regime probed by present day inelastic neutron scattering, no propagating sound waves have been observed in some quite simple systems such as SF_6 . It would be desirable to extend these measurements to the smaller Q 's and ω 's where the phonons can propagate. This again necessitates higher fluxes in order to "squeeze down" the energy and momentum resolution.

The above are just a small selection of the many problems in the study of collective excitations that require more intensity. It is fair to say that a considerable gain could be achieved on existing instruments with improved monochromators and beam optics. However, these improvements should also be considered during the design of any new source. They would make the new instruments even more powerful.

3. Instruments Needed

3.1 The Triple Axis Spectrometer

For studies of collective excitations localized in wavevector and energy, the triple axis spectrometer is the present instrument of choice⁽¹⁾. The principal advantages are: 1) it concentrates its spectral resolution on a specific region of Q and ω ; 2) it permits rapid variation of external scan variables such as temperature, field or pressure, made at a single Q, ω value; 3) its resolution, as well as its energy and wavevector range, may be easily and rapidly changed; 4) the data as measured are immediately understandable, being proportional to $S(Q, \omega)$.

It will likely remain the prime instrument for forefront condensed-matter research where a search is being carried out for a new phenomenon of unknown cross-section. We consider that experiments where the cross-section is known ahead of time are seldom at the leading edge of investigative science. The triple-axis spectrometer requires a steady state source. Signal gain factors for a new high-flux reactor over present U.S. capability amount to five from the flux, approximately $2\frac{1}{2}$ from beam tubes and 4 from improved vertical focussing, with the improved beam optics which a new source and matching instruments would allow. Backgrounds will remain in the range 0.1 -1 cpm. For very low signal experiments the fast neutron background could be eliminated at a 50% cost in count rate by the use of a simple chopper and gating system as is common in optical spectroscopy.

3.2 Time-of-Flight Crystal Analyzer Instruments

There are currently three instruments specifically designed for the study of collective excitations on pulsed neutron sources: Constant- Q Spectrometers⁽⁵⁾ at Harwell, England and Los Alamos and a High Symmetry Spectrometer (MAX) at KENS, Japan⁽⁶⁾. The two geometrics are shown in Figure 1. Both instruments are deliberately constrained to take data on a line in reciprocal space parallel to $\underline{k}_0$, for a single setting of sample and analyzer. In other words Q_1 , the component of Q perpendicular to $\underline{k}_0$, is held constant for all data collected at that setting

and the intensity is distributed over the (Q_{11}, ω) plane where Q_{11} is the component of Q parallel to k_0 . In the High Symmetry case, from the sample onwards the instrument is similar to a reactor triple-axis spectrometer with a large number of analyzers, each with its own collimators and detector. Figure 2 shows the layout and some early results from the MAX spectrometer. In the Constant-Q Spectrometer a large extended analyzer of one fixed orientation is used in conjunction with an area detector or continuous array of detectors. This has the advantage that data are collected over a fine mesh in (Q, ω) space and that it can be rebinned or interpolated to give any scan in the (Q_{11}, ω) plane, including Constant-Q scans. The price to be paid, however, is that Q_1 can no longer be tuned to a specific value and that a lot of the information collected will, in general, be in off-symmetry directions and may not be useful.

The Constant-Q Spectrometer at Los Alamos has recently been fitted with a 25 cm long pressed germanium analyzer and data sets demonstrating phonon dispersion in aluminum have been collected⁽¹⁰⁾. The equivalent time integrated neutron background is better than 0.2 cts/min/detector, but that figure is not precise enough for use in estimating background levels on the proposed new pulsed source. While the neutron background can definitely be reduced further with the use of better shielding around the analyzer and by placing collimators before and after the analyzer, we suspect that backgrounds may be higher than on chopper spectrometers, simply because the full range of wavelengths, in the white incident beam, is incident on the sample.

A number of measurements have been made on the MAX spectrometer. These data have been analyzed as time-of-flight scans and the peak positions plotted in (Q_{11}, ω) space. The analyzers are currently positioned at a 4° angular spacing. The (Q_{11}, ω) mesh could be made sufficiently fine for the construction of Constant-Q or Constant- ω scans if the analyzers could be placed at a spacing of 0.5° . While difficult, this is possible from an engineering point of view and there are plans to build such an instrument at Rutherford Lab, U.K.

The scans made on the pulsed source instruments are different from conventional triple-axis constant-Q scans. Neither E_0 nor E_1 are kept constant. Instead, the crystal orientation is kept constant. The pulsed source instruments are useful for Type A collective excitation experiments provided data at one crystal setting can be collected within short times of order a few hours. Subject to similar constraints it is also possible to conduct several simultaneous constant-Q scans on a triple-axis spectrometer equipped with several analyzers. Indeed one could also construct a steady-state instrument in either the Constant-Q

or High Symmetry Configurations that scan incident energy E_0 by means of angular increments on a crystal monochromator, rather than by means of time-of-flight as used on the pulsed sources. Figure 3 sketches the use of such a steady-state High Symmetry Spectrometer. For this instrument

- 1) the incident energy must be stepped to obtain a complete constant-Q scan.
- 2) adjacent analyzers can be arranged to assemble constant-Q data along a radial or transverse symmetry line.
- 3) the incident as well as the scattered resolution may easily be changed.
- 4) the open geometry will likely lead to higher backgrounds.
- 5) the constraints on relative $h\omega$ step length for adjacent analyzers provide a limit to the instrument's utility.

The resolution properties of the pulsed source instruments have been discussed before by Windsor et al.(7) and Pynn and Robinson(8). While it is easy to change the resolution of the scattered beam, there is less flexibility in the incident energy resolution. This is determined principally by the time pulse width and the incident flight path, neither of which can be changed easily. One can however consider changing the intensity and pulse width at the moderator by moving the poisoning sheet within a liquid moderator or by flooding cavities on the front surface, as shown in Figure 4. In any case, the incident energy resolution is normally rather good. One can use the same focussing techniques as on reactors, but longitudinal phonons will be easier to focus than transverse phonons, in contrast to the reactor conditions.

3.3 Chopper Spectrometers

The other means of energy analysis normally used at reactors is to produce a pulsed monochromatic beam with a pair of phased choppers or rotors. The same technique is well suited to the pulsed source, except that one needs only one chopper which must be phased to the accelerator. In the early days of neutron scattering this technique was used occasionally in the measurement of dispersion curves, but it has since fallen out of favor. There are two reasons for this. Firstly, the quantity of information collected is large and a significant amount of computation is necessary to display or analyze it. Much of Q, ω space is devoid of significant information for some systems. Given the present state of computer technology and its rate of advance, that can no longer be considered a real objection. More fundamental is the fact that these machines collect data on a paraboloid of revolution in (Q, ω) space, rather than on a plane, as in the crystal analyzer methods, or point by point as in the triple axis case. There are great

conceptual difficulties in visualizing scattered intensity distributed over this surface, unless there is another symmetry in the problem. For instance, if the scattering is dispersionless or the dispersion surface is isotropic, this type of machine is useful, as illustrated by the recent measurement of spin wave dispersion of Fe at IPNS.(4)

The chopper instruments become most productive for experiments where the scattering of interest extends over the whole plane of the Brillouin zone. A wide range of times are measured by scans with numerous counters can all provide useful information. By rotating the sample over the irreducible quadrant of the scattering plane an $S(Q, \omega)$ map is obtained, providing a highly detailed description of the scattering law over many zones.

4. General Comparisons of Instruments on a Reactor and on a Pulse Source

For experiments of type A where only one Q, ω value is of interest we may make a comparison independent of the details of instrument configuration and resolution by assuming that either instrument picks out the same incident and scattered energy window and solid angle. The performance then becomes simply proportional to the time-averaged flux of the source. This makes performance on the projected pulse source lower than that on the reactor by a factor of order 50. In fact it varies from 70 at 1 meV to 20 at 500 meV. Of course, for energies above 50 meV, the poor monochromator reflectivity must be taken into account on the reactor source. The only other major error in this approach is that a crystal monochromator gives good angular resolution when defining the energy window, whereas the two are decoupled for the pulsed source.

Experiments of type B require a scan either in ω or in Q along a defined direction or in both. A multiple analyzer on a pulsed source is able to perform an energy scan at constant Q but this is not possible on a reactor multianalyzer with more than two arms. Constant Q configurations therefore gain over the mean flux comparison by the number of counters contributing to the constant Q scan. This may be typically 20 depending on the length of the ω scan needed and the angular spacing of the multiscanner. The reactor to pulsed performance ratio is therefore 20/50, so the two sources become roughly comparable in count-rate for constant Q scans. However the Constant Q Spectrometer configuration has important limitations in flexibility compared to a triple axis on a reactor.

If the range of type B experiments is widened to include a scan in Q, ω along a defined direction in reciprocal space, experiments can be performed on either a reactor or a pulsed source by using a multi-analyzer. The number of simultaneous

Q vectors possible is equal to the number of analyzers, say 10 in the triple axis case. In the pulsed case we have a similar number defining the ω steps but the Q scan is essentially continuous and equal to the number of useful resolution widths along the time-of-flight scan. We suggest a figure around 40 depending on the breadth of the useful ω scan along the chosen Q direction. Thus the multi-analyzer factor cancels out for reactor and pulsed multi-analyzer instruments and the pulsed instruments gain over the mean flux figure by a factor 40 making them roughly comparable considering the 1/50 mean flux differential. However the multi-analyzer on a reactor has more flexibility than the same configuration on a pulsed source. In particular it can perform scans where the reduced q -vector is parallel or perpendicular to Q .

Class C experiments seek to measure continuous distributions of $S(Q, \omega)$ over a whole plane of reciprocal space. Phased chopper instruments on both reactor and pulsed sources are ideal for this task. 50 counters may be usefully employed together say 100 of useful resolution elements in the time-of-flight scan. A gain factor of some 5000 over the mean flux is therefore feasible putting the proposed new pulse source a factor 100 ahead of the reactor. Another way of looking at the comparison is to say that pulsed source choppers see the "peak flux" of the source. Another important advantage of rotor spectrometers is that the Q resolution may be decoupled from the energy resolution. This is not possible in either reactor triple axis or pulsed-source-crystal-analyzer instruments. On the other hand a disadvantage of rotor spectrometers is that the energy resolution cannot conveniently be changed to suit the experiment.

The results are summarized in table 1. The triple axis is pre-eminent in classes A and B(i), with the multi-analyzer looking very promising on either source for B(ii). The pulsed rotor is pre-eminent for C.

For comparisons more detailed than these, it is necessary to examine particular experiments and this is done in the next section.

Table 1

Approximate Gain Factors for Collective Excitation Instruments

The table gives for thermal energies counters x time elements x mean flux		A	B(1)	B(11)	C
		Q defined ω defined	Q defined ω scanned	Q on a line ω scanned	Q scanned ω scanned
Reactor proposal	Triple axis	1 x 1	1 x 1	1 x 1	
	Triple axis + multi-analyzer			10 x 1	
Pulsed proposal	Single analyzer	1 x 1/50			
	CQS		20 x 1/50	20 x 1/50	
	Multi-analyzer			10 x 40/50	10 x 100/50
	Rotor				50 x 100/50

5. Detailed Flux Estimates Made Using the Program SIP

The Shelter Island Program (SIP) is briefly described in an appendix. It was used in two modes. The first was a benchmark mode reviewing experiments already performed. The experiments were (i) Phonons in $\text{NbD}_{0.85}$ on a Brookhaven triple axis, (ii) Magnons in Fe on the Argonne chopper. For these experiments, the instrumental parameters were mostly on hand and it was possible to arrive at sample fluxes and count-rates which could be at least roughly checked with experiment. The second mode explored the performance of the two new sources for experiments difficult or impossible on today's sources. The desire was to obtain a count-rate above likely background for a specified Q, ω and resolution, leaving the program to choose the best experimental configurations given the desired resolution.

(i) Longitudinal acoustic pPhonon measurements on a $\text{NbD}_{0.85}$ single crystal.

$$Q = 3.85 \text{ \AA}^{-1}, W = 7 \text{ meV}, E_1 = 14.7 \text{ meV}$$

Brookhaven HFBR triple axis

Monochromator and analyzer PG(002) $d=3.3514 \text{ \AA}$, $\eta=0.4^\circ$, Reflectivity = 0.7

Collimations $\alpha_1 = 0.67^\circ$, $\alpha_0 = 0.33^\circ$, $\alpha_1 = 0.67^\circ$, $\alpha_2 = 0.67^\circ$ (no focussing)

$$\beta_1 = 0.5^\circ, \beta_0 = 2.2^\circ, \beta_1 = 3.8^\circ, \beta_2 = 5.7^\circ \text{ (focussing x 2)}$$

Sample Size width = 2.5 cm, height = 2.5 cm, thickness = 2.5 cm

Macroscopic cross-section integrated over phonon peak

$$\frac{\sigma_{\text{coh}}}{4\pi} \cdot \frac{N_A \cdot \rho \cdot t}{M} \cdot \frac{k_1}{k_0} \cdot \frac{\hbar^2 Q^2}{2M \cdot \hbar \omega_0} \cdot e^{kT/\hbar \omega}$$

$$= \frac{6.24}{4\pi} \cdot \frac{0.6025 \cdot 8.6 \times 2.5}{92.91} \cdot \frac{2.66}{3.00} \cdot \frac{14.88}{92.91 \times 4} \cdot \frac{25.8}{4} = 0.0158$$

The results were	measured	actual calculation
Flux at moderator surface	-	$1.1 \times 10^{15} \text{ cm}^{-2}\text{s}^{-1}\text{sterad}^{-1}\text{eV}^{-1}$
Flux at monochromator surface	$4.10^9 \text{ cm}^{-2}\text{s}^{-1}$	$8 \times 10^9 \text{ cm}^{-2}\text{s}^{-1}$
Flux at sample	$1.4 \times 10^7 \text{ cm}^{-2}\text{s}^{-1}$	$2.1 \times 10^7 \text{ n cm}^{-2}\text{s}^{-1}$
Count rate	252 cts/min	168 cts min

(ii) Magnon measurements on an Fe single crystal⁽⁴⁾

$Q = 4.86 \text{ \AA}^{-1}$, $W = 65 \text{ meV}$, $E_0 = 200 \text{ meV}$

Argonne LRMECS instrument

Rotor parameters Peripheral velocity $0.0127 \text{ cm } \mu\text{s}^{-1}$, diameter 15 cm,

Absorber thickness 0.025 cm, slit width 0.1 cm

Incident flight path $L_0 = 1350 \text{ cm}$, $\alpha_0 = 0.43^\circ$, $\beta_0 = 0.43^\circ$

Scattered flight path $L_1 = 250 \text{ cm}$, $\alpha_1 = 0.57^\circ$, $\beta_1 = 5^\circ$

Sample size ~ 2 cm width ~ 2 cm height, ~ 2 cm thickness

Sample cross-section

$$\frac{N_A \rho t}{M} \cdot \frac{k_1}{k_0} (r_0 \gamma)^2 \cdot F(Q) \cdot \frac{S}{2}$$

$$= \frac{0.6025 \times 7.86}{50} \times 0.906 \times 0.29 \times \frac{5}{2} \times 0.5 = 0.055$$

	measured	calculated
Giving		
Flux at moderator surface		$2.09 \times 10^{10} \text{ cm}^{-2}\text{s}^{-1}\text{eV}^{-1}\text{sterad}^{-1}$
Flux at sample	602	$919 \text{ n cm}^{-2}\text{s}^{-1}$
Count rate	4.3 cts/hr	3.6 cts/hr

We next consider the count-rate computed for both new sources on different instruments for experiments which are difficult or impossible on present day sources. Their feasibility depends on whether they will give adequate intensity

above background estimates. Unfortunately backgrounds on the new sources are very difficult to estimate and we give present day values. Two experiments are chosen for this study.

- (i) Near quasi-elastic scattering with very high resolution.

$$Q = 1.33 \text{ \AA}^{-1}, \Delta Q = 0.01 \text{ \AA}^{-1}, \omega \sim 0, \Delta\omega = 0.4 \text{ meV}, E_0 = 14.7 \text{ meV}.$$

This configuration is of very wide applicability in triple axis work. The actual specification is from a recent experiment on dilute magnets, but it is equally applicable to gap spectroscopy in semiconductors or low energy solitons resolvable from an elastic peak.

- (ii) Inelastic scattering at low cross-sections.

$$Q = 4.86 \text{ \AA}^{-1}, \Delta Q = 0.1 \text{ \AA}^{-1}, \omega = 65 \text{ meV}, \Delta\omega = 5 \text{ meV}, E_0 = 200 \text{ meV}.$$

This configuration is recognizable as the Fe magnon considered earlier. Here we consider it on the new sources but with a much lower cross-section $10^{-4} \text{ bn sterad}^{-1} \text{ eV}^{-1}$ corresponding to continuous Stoner mode scattering.

Both problems are considered on three instruments.

- (a) A triple axis on HFIR II with horizontal collimations chosen to give the desired resolution, and related vertical collimation possible with a 15 cm high source and focussing to a 2cm sample. Otherwise the instrument is a standard HFBR design.
- (b) A crystal analyzer on ASPUN with an analyzer identical to the one above. The planes are PG(002) at 14.7 meV and Ge(220) at 135 meV. The initial flight path is chosen to give the required resolution.
- (c) A rotor spectrometer on ASPUN. The actual rotor parameters are those in use at Argonne. The initial and final flight path are chosen by the program to satisfy the resolution.

	HFIR II Triple axis	ASPUN	
		crystal analyzer	rotor time-of-flight
Background?	0.1 - 1 cts m^{-1}	0.1 cts hr^{-1}	< 0.1 cts hr^{-1}
(i) High resolution quasi-elastic	0.45 cts m^{-1}	0.45 cts h^{-1}	6×10^{-4} cts hr^{-1}
(ii) Low cross-section inelastic	1.23 cts m^{-1}	0.32 cts h^{-1}	0.22 cts h^{-1}

Thus there appears a rough equivalence in count rate for a single resolution element (publishable point) between counts per minute on the new reactor and counts per hour on the new pulsed source. This reflects the ratio in the mean flux between the sources. Since the likely background also follows this ratio it follows that the signal to background is comparable for these experiments, and all appear possible with the exception of the rotor spectrometer used for the very high resolution experiment. The performance difference for a complete experiment will depend on the number of simultaneous time elements and counters as described by Table 1.

5. Benchmark Experiments

There has been ample experience with triple-axis instruments in this country at the various reactor centers, but there has been little quantitative inter-comparison of the performance of these instruments. On pulsed-sources the crystal analyzer instruments may be competitive with the 3-axis instruments. Two types have been constructed but there has been little opportunity to gain experience so far.

It is proposed that an effort be made to compare measurements on a standard sample that is circulated from laboratory to laboratory where the experimental conditions are well defined and the flux on the sample properly measured. The sample could be a single crystal of niobium where the phonons are well known and energies of the acoustic modes can be measured near 5 meV transfer as well as 20 meV. The measured count rates could then be compared with those calculated. In this way one could experimentally evaluate the merits of triple-axis instruments on a steady state source and the crystal analyzer instruments at pulsed sources.

6. Conclusions

The most challenging experiments will be ones which require a study of only a limited region of (Q, ω) space. These are best done with a triple axis crystal spectrometer whose intensity depends on the average flux, which is clearly higher for a steady-state neutron source. In this regard a HFIR-II source would be particularly useful.

Surveys along a line in reciprocal space may be efficiently performed on several instruments with multiple detectors and one or more analyzers. It is unclear whether these instruments are best placed on a pulsed or steady-state neutron source. We suggest that there be more investigation of these methods, particularly at reactor sources, to see if they can be used to produce data more quickly for problems which require data over wide ranges at Q and ω .

Broad surveys over all of Q and ω are best performed with conventional rotor time-of-flight techniques and the pulsed source is likely to produce the better flux.

We suggest an exchange of samples, possibly Nb, on which typical phonons can be measured with different instruments so as to be able to compare the results with calculations of the flux in detail.

Appendix - SIP - The Shelter Island Program for Exploring Neutron Scattering Triangles, Resolutions and Intensities

The two goals of the Shelter Island Workshop were to explore what science may be possible on the new sources and to compare their performance. In practical terms this means deciding on the Q, ω range, resolution and cross-section of chosen experiments and estimating a count-rate to compare with a background estimate - the calculation to be done on both sources. SIP is designed to do exactly that for inelastic scattering, in particular for the triple axis spectrometer on reactors, the time-of-flight crystal analyzer, and the rotor and crystal monochromator time-of-flight instruments. The program works simultaneously in two modes. The "Actual" mode requires the instrument configuration to be specified and will evaluate count-rates at the sample and detector for a given source, sample size and cross-section. The actual Q and ω resolution is also estimated. In the "Best" mode the resolutions ΔQ and $\Delta \omega$ must be specified. Using simple matching arguments, which neglect focussing, it decides on a spectrometer configuration with collimations and flight paths which will achieve the desired resolutions. The "best" count-rates may be compared with the actual values.

The program includes a data base of 6 reactors and 6 pulsed sources worldwide, with a parameterization of their flux distribution and pulse width (FWH). Both include Maxwellian and epithermal terms. The fluxes are always specified in the conventional way, i.e. the energy-integrated Maxwellian flux per $\text{cm}^{-2}\text{s}^{-1}$ and the epithermal flux from the whole moderator surface at 1 eV per $\text{s}^{-1}\text{sterad}^{-1}\text{eV}^{-1}$.

$$MF = \frac{M_{TF}}{4\pi} \cdot \frac{E_0 \times 1000}{E_{kT}^2} e^{-E_0/kT} + \frac{N_{EP}}{(E_0/1000)^{1-BUC}} \cdot \frac{WJF}{M_{HI} \cdot M_{WI}}$$

where

$$WJF = \frac{1 + (5 E_{kT}/1000)^7}{1 + (5 E_{kT}/E_0)^7}$$

The pulse width is given by

$$PW = \left\{ SPW^2 + \left(MWW \times \sqrt{\frac{33.54}{E_{kT}}} \cdot MJF \right)^2 + \left(\frac{EPI}{(E_0/1000)^{\frac{1}{2}}} (1-MJF) \right)^2 \right\}^{\frac{1}{2}}$$

where

$$MJF = \left(1 + \frac{E_0}{E_{kT}}\right) e^{-E_0/E_{kT}}$$

All the symbols and their units are defined in table A. WJF is the familiar Westcott joining function. MJF is the integral of the Maxwellian flux from infinite energy down to E_0 . This formula gives an excellent fit to the curves for pulse width in Appendix 1 of this report.

Irrespective of the instrument, the sample flux SF is evaluated as the moderator flux $s^{-1}sterad^{-1}eV^{-1}cm^{-2}$ times a "monochromator element" ME describing the product of the incident solid angle $\Delta\Omega_0$ and incident energy window ΔE_0 . It therefore has units sterad eV and gives a sample flux in $cm^{-2}s^{-1}$ units. For example a crystal monochromator in the conventional notation is

$$ME = \frac{2E_0}{1000} \times \cot\theta \times \frac{\alpha_I \alpha_O \eta_M}{\left(\alpha_I^2 + \alpha_O^2 + 4\eta_M^2\right)^{\frac{1}{2}}} \\ \times \frac{\beta_I \beta_O}{\left(\beta_I^2 + \beta_O^2 + 4\sin^2\theta_M \eta_M^2\right)^{\frac{1}{2}}} \times \text{Reflectivity}$$

The "best" value for this element for incident resolution $R_0 = \Delta k_0/k_0$ is

$$ME^B = \frac{2E_0}{1000} \tan\theta_M R_0^2 \beta_I \times \text{reflectivity}$$

The final count-rate of a triple axis will be a product of the moderator flux MF, the monochromator element ME, a sample element SE equal to the sample area times its macroscopic cross-section per unit area in $sterad^{-1}eV^{-1}$ units, an analyzer element AE exactly as for the monochromator, and a counter efficiency element CE.

$$CR = MF.ME.CE.AE.CE.$$

For time-of-flight elements in either incident or scattered beam we evaluate the counts per "resolution element". This is roughly the counts integrated over a resolution broadened peak in the time-of-flight spectrum. This must take into account the time uncertainty introduced by the other element. For example, in a time-of-flight crystal analyzer instrument the time uncertainty introduced by the analyzer path is evaluated first, so that it may be included with the source pulse width and channel width in evaluating the incident beam resolution.

Many time-of-flight instruments use several "resolution elements" along the time-of-flight scan and possibly over many counters. The number of resolution elements must be estimated separately and used to multiply the count rate given by the program before comparison with say a single counter triple axis count-rate.

The program operates interactively and is written in standard IBM PC BASIC of some 5600 lines.

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Table A
A Selection from SIP Source Database

<u>Parameter</u>	<u>Symbol</u>	<u>Unit</u>	<u>BNL</u>	<u>HFIR II</u>	<u>IPNS</u>	<u>ASPUN "A"</u>
Maxwellian total flux	MTF	$s^{-1}cm^{-2}$	10^{15}	5.10^{15}	2.10^{11}	1.310^{14}
Moderator width	MWI	cm	10 ?	10 ?	10	10
Moderator height	MHI	cm	10 ?	15 ?	10	10
Number epithermal neutrons at 1eV	NEP	$sterad^{-1}eV^{-1}s^{-1}$	3.2×10^{14}	1.6×10^{15}	4.5×10^{11}	4×10^{14}
Buckling	BUC		0	0	0.15	0.15
Source pulse width	SPN	μs	-	-	0.2	0.2
Epithermal pulse width at 1 eV	EPI	μs	-	-	2	2
Maxwellian width (amb, $E_0 \rightarrow 0$)	MWW	μs	-	-	36.2	50
Thermal energy (amb)	EKT	meV	25.8	25.8	33.5	33.5

Fig. 1. Crystal analyzer spectrometers (i) Multi-Analyzer Crystal (MAX) design has independent analyzers and can direct the time-of-flight scan down a desired line in reciprocal space. (ii) The Constant Q Spectrometer (CQS) design can perform an energy scan at a specified Q. Only certain time channels from each counter are included in the scan.

Fig. 2. The MAX spectrometer in Japan and some of its results.

Fig. 3. The multi-analyzer instrument on a reactor. Constant Q energy scans are not possible but each analyzer can be directed to a selected Q point and the frequency scanned by sequential changes in E_0 . The reactor instrument has the advantage that the crystal angle α may be changed during the scan. For useful ω scans the analyzers need to be close together and independently driven.

Fig. 4. Possible arrangements of a water moderator with cadmium poisoning which would enable the incident resolution to be varied.

Crystallography (Powder)

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A. Introduction

The use of neutron powder diffraction for both basic and applied research has increased dramatically over the last decade. At facilities which offer powder diffraction capability to outside users, the requests for instrument time consistently exceed availability. This renewed interest in powder diffraction is the result of the design and construction of high-resolution, high-intensity powder diffractometers and the development of data analysis methods, such as Rietveld structural refinement, which allow precise structural information to be obtained from powder data. Neutron powder diffraction is also finding new uses in applied materials science (e.g., the measurement of residual stress in technologically important materials) and chemistry (e.g., time-resolved studies of chemical reactions).

The new, high-flux neutron sources (i.e., 10^{16} n-cm⁻²-s⁻¹ reactor or 10^{17} n-cm⁻²-s⁻¹ peak flux pulsed source) will allow powder diffractometer performance to be even further extended and open some new scientific areas. At these flux levels, the resolution of neutron powder diffractometers is expected to reach $\Delta d/d = 3 \times 10^{-4}$. This is comparable to high resolution x-ray diffractometers including those currently being tested at synchrotron light sources. Additionally, the neutron diffraction method offers the natural advantages of well-defined instrumental line shapes, constant scattering cross sections (no form-factor), penetrating power, and freedom from texture and preferred orientation which are often associated with small x-ray samples. Thus, neutron powder diffraction is expected to be the preferred method for many kinds of problems.

B. Scientific Opportunities

Based on current experience and the projected performance of future instruments, neutron powder diffraction is expected to play a major role in four general scientific areas over the next decade:

(1) Structure Solution and Refinement

Many materials are simply not available as single crystals, yet knowledge of their crystal structure is basic to an understanding of their physical and chemical properties. Catalysts are a typical example of such "real" materials. In their active state they are finely divided. Even if single crystals can be grown, crystal growth may itself modify their stoichiometry, or reduce their activity. Examples are heavy metal oxides and zeolites. There are, as well, examples of catalysts which may exist as multiple phases, or which are not really thermodynamically stable. Crystal growth is of course a method of purification and selection of the most stable phase; this is often not what is wanted.

Other real materials are prepared only as ceramics - whether or not single crystals can be grown may not be of interest, or may be an unnecessary complication if the crystal structure can be determined by other means. Again, crystals may be multidomain, as in certain dielectric and magnetic materials, so that single crystal techniques are difficult or impossible. Finally, materials which crystallize only under special conditions of low temperature, high pressure, sublimation, sputtering, solid state reaction etc., may be impossible to prepare as single crystals.

It is not surprising that these real materials, which cannot be prepared as single crystals, seem to be a growing proportion of all materials studied by neutron crystallography. Neutron powder diffraction has in the past ten years seen rapidly growing applications in this area. High resolution neutron powder diffraction, with new computerized "information extraction" techniques, has been shown to produce structural information of a precision approaching that of single crystal techniques in many cases, and much better than that obtained by present x-ray powder methods.

To date, neutron powder diffraction has been mainly used to refine models of the crystal structure. However, the eventual goal is to directly solve the structure using powder data alone. This is possible, but only with very high resolution diffractometers. Thus, the growing complexity of the structural problems attempted inevitably points to the need for diffractometers of higher resolution.

Higher resolution means, in practice, higher intensity neutron sources. If the dimensions of the crystal unit cell are doubled, the number of diffraction lines increases by a factor of eight, and their average intensity decreases by a factor of eight. Improving the resolution of the diffractometer by the same factor, unavoidably results in a loss of intensity which often cannot be com-

compensated for by the use of multidetectors, focussing or other instrumental techniques. Thus, a modest increase in the size of the problem implies a substantial increase in the required neutron intensity. Some gains are possible by optimizing diffractometer design, but there is no escaping the inevitable conclusion - higher flux neutron sources are needed.

With the presently operating neutron powder diffractometers such as D1A at the ILL, GPPD at IPNS, 2XD at the University of Missouri Research Reactor (MURR) or BT1 at NBS, the largest number of structural parameters that can be refined is about 100. New instruments which will shortly come into operation, D2B at ILL and HRPD at the SNS (Rutherford Appleton Lab.), will offer resolution about four times better than that which is currently available and are expected to extend this limit to about 300 parameters in favorable case where sample-dependent line broadening is not present. Fluxes an order of magnitude higher, as presently proposed for both reactors and pulsed neutron sources, would make possible an additional improvement in resolution and could extend this limit to around 500 parameters.

2. Time-Resolved Studies

The application of neutron powder techniques to real time diffraction studies has been limited to date, undoubtedly because of the comparatively slow data collection rates at most constant-wavelength (CW) instruments, typically several hours. Many of these instruments are optimized for high resolution, whereas, for time-resolved studies, it is advantageous to have moderate resolution with a large-area detector. This kind of instrument is exemplified by D1B at ILL, which covers a 2θ range of 80° and has a resolution, $\Delta d/d$, of about 1% at the focussing position ($2\theta \approx 44^\circ$) with a sample flux of $2 \times 10^6 \text{ n-cm}^{-2}\text{-s}^{-1}$. This permits complete data to be collected in 1-10 minutes in many cases, and has been used extensively for studies of chemical reactions, phase transformation kinetics and crystallization processes. TOF techniques are also well-suited for real time studies, and useful data can be collected on the Special Environment Powder Diffractometer at IPNS in as little as 5-10 minutes in some cases.

A new instrument, D20, is nearing completion at ILL and will be a greatly improved version of D1B, with an angular range of 160° , resolution of 0.5% at the focussing minimum and a flux on the sample of about $10^8 \text{ n-cm}^{-2}\text{-s}^{-1}$. This will permit complete data to be collected in a few seconds, and will also allow "time-slices" of about 1 msec for experiments involving repetitive measurements in which the detector is phased to some pulsed external variable such as a magnetic

field or a laser flash. This will provide a means of studying periodically induced transient phenomena such as magnetic relaxation or short-lived reaction species. Continued development of this type of instrument at a more intense source could eventually lead to an order of magnitude more sample flux and a substantially larger useful sample volume, resulting in three orders of magnitude improvement over the existing machine, D1B.

It is not easy at this stage to predict the impact that such an instrument would have in the general area of time-resolved studies, since the full potential of the technique is far from being realized on present day sources. However, it is clear that there are wide-ranging applications in chemistry, solid state physics, ceramics, metallurgy, polymers and possibly biological systems. A partial list would include chemical reaction kinetics, intermediate species, rapid phase equilibria studies, identification of new unstable compounds, in-situ studies of sputtering and chemical vapor deposition, crystallization and grain growth studies, annealing, precipitation, order-disorder kinetics, diffusion at solid interfaces and hydration-dehydration kinetics. Furthermore, the comparative ease of doing these experiments in controlled environments, e.g., furnaces, high pressure cells, inert and reactive atmospheres, is a very attractive feature of the neutron powder technique in comparison with x-ray diffraction methods.

3. Small Samples

A number of important experiments with small samples (or small accessible sample volumes) become possible with the higher neutron fluxes which will be available at future sources. Present experience indicates that with existing instruments (in the most favorable circumstances), roughly 0.1 g is the lower limit for high resolution neutron powder diffraction while 0.01 g is the limit for low resolution powder diffraction. With the new high fluxes combined with further advances in instrument design it should be possible to perform high resolution powder diffraction measurements on samples as small as 1 mg. This will make possible the study of samples which can be produced only in small quantities by techniques such as chemical vapor deposition or sputtering. Compounds which contain exotic or very expensive elements (e.g., transuranics) or particular isotopes which are available in very limited quantities will also become possible to study. Very small samples can also be used to overcome the problems of doing diffraction studies of compounds containing elements with high neutron absorption cross sections (e.g., Gd, Dy, Cd, Eu).

Another important class of "small sample" problems are those in which the volume of interest or the accessible volume of the sample is small. The first category includes the use of neutron diffraction as a nondestructive probe of a localized volume of a larger sample (as discussed in the following section on mechanical properties). Diffraction in a high pressure environment is an example of the second case.

High pressure neutron diffraction implies a small accessible sample volume because of the limitations of the materials used to enclose samples under pressure. The range of devices currently in use extends from hollow-cylinder, gas or liquid cells which enclose large volumes of several cm^3 at pressures up to about 10 kbar to the recently popularized diamond anvil cells which achieve pressures in the Mbar range for samples with dimensions of 100 μm or less. Each increase in pressure range is accompanied by an order of magnitude reduction in sample size. Neutron diffraction studies are currently flux limited and can be done for 1 cm^3 samples in gas or liquid cells to 10 kbar or 0.1 cm^3 samples in supported piston-in-cylinder cells which can reach about 40 kbar. The next order of magnitude increase in neutron flux will allow the use of multiple anvil (cubic) presses which can achieve 100 kbar for effective sample volumes (accessible to neutrons) of about 0.01 cm^3 . With still higher fluxes, opposed anvil designs similar to diamond anvil cells may extend this range to a few hundred kbar.

Physical problems of interest at high pressure included both structural and electronically-driven phase transitions, high-pressure reaction products, geophysical studies and the correlation of structural changes with other physical properties which vary with pressure. An example in the last category is the sharp pressure dependence of the superconducting transition temperature versus pressure for many high- T_c , ternary superconductors. High pressure diffraction studies could help establish the relationship between structure and superconductivity in these materials. An interesting example of phase transitions is the sequence of ordering transitions observed as a function of pressure in intercalation compounds. In these and many other systems the range of novel behavior that can be observed is far greater as a function of pressure than temperature.

4. Applied Materials Science - Mechanical Properties

The neutron has some significant advantages as a probe of the mechanical/metallurgical properties of many systems because of its high penetrating power compared to x-rays. In particular, the last few years have seen considerable activity at almost all neutron scattering centers in the measurement

of texture and stress. In the case of texture, there are numerous systems in which the surface texture is different from that of the bulk. The ability of the neutron to penetrate, provides a more accurate representation of the bulk properties than any other non-destructive probe. In addition the generally large volume probed by the neutron leads to a reliable statistical average over the bulk. Two methods are available for these studies. One uses the intensity of a given Bragg peak as a function of direction in the sample to determine a grain distribution function (pole figure). The other uses more traditional powder methods to compare the relative Bragg intensities of a group of reflections from which the distribution is deduced. The measurements tend to be slow when many adjacent volume elements are probed to provide information about homogeneity and processing technology. Clearly these measurements benefit from both higher fluxes and more efficient detection capability. A more stringent requirement is imposed on the instrument in the determination of the strain tensor within a sample (residual or applied stress). These measurements examine the shift of one or more Bragg reflections from the position observed for an unstressed case, as the sample axis is varied (rotations about the beam) and as a small volume element defined by the intersection of incoming and outgoing beam paths is moved through the sample (or the sample is moved across the volume element). From the changes in position of the Bragg reflections the three-dimensional strain tensor can be deduced. Because x-ray beams cannot penetrate substantially below the surface in most materials of interest, such measurements have never been performed, and only surface stresses (2-dimensional) have been determined, with x-rays. However, such measurements have recently been carried out with neutrons both on idealized specimens (bent beams, cylinders in tension or compression, etc.) and on actual service pieces (welded sections) and are quite informative. The experiments require both good resolution, to accurately define $\Delta d/d$, and high intensities, in order to reduce the probe volume to regions of physical interest, as in the case of welds, rather than to average over volumes which are large compared to the stress gradients. Accuracy in the definition of the peak shape is also of key importance for this application, and is especially important in those cases in which not only is the mean strain determined from the peak position, but the stress distribution is also deduced from the peak breadth.

Narrow peaks and accurate knowledge of the instrumental resolution also allows one to extract information about the particle size and strain distribution from the angular or wavelength dependence of the peak breadths in a diffraction diagram. As resolution increases, this application and the relation of these

parameters to other properties of the system, such as chemical activity, will undoubtedly be more thoroughly examined.

The proposed higher neutron fluxes are expected to allow the probed volume to be reduced to about 1 mm^3 . Since neutron powder peak shapes are well understood and can be accurately parameterized, peak positions (for a single peak) can typically be determined to a precision one to two orders of magnitude better than the diffractometer resolution (FWHM).

C. Instrumentation

Well-designed powder diffractometers have been operated at both reactors and pulsed neutron sources for a number of years. This has provided a considerable amount of data on relative instrument performance and allows the capabilities of future instrument designs to be evaluated with some confidence. For the purpose of evaluating various instrument designs, the performance parameters of existing powder diffractometers will be discussed and criteria developed for making meaningful comparisons. These criteria will then be applied to instruments now under construction or about to begin operation and to proposed designs for a $10^{16} \text{ n-cm}^{-2}\text{-s}^{-1}$ reactor or a $10^{17} \text{ n-cm}^{-2}\text{-s}^{-1}$ pulsed source. The instruments included in the discussion are admittedly a small sample of those currently in operation but do adequately represent the present state-of-the-art.

1. Present-Day Instruments

In this section, the design features and performance characteristics of instruments at various existing neutron centers are described. Some general details of these are provided in Table 1. These instruments have been operational for some time and considerable experience has been gained. D1A and GPPD are designed principally for high resolution studies with a corresponding sacrifice of intensity, while D1B is designed for high intensity experiments at the expense of resolution. 2XD represents a compromise in these two respects.

Some of the basic operational parameters of these machines are summarized in Table 2. The various quantities listed are defined as follows:

- (i) λ is the normal operating wavelength for the CW instruments.
- (ii) $Q_{\text{max}} = 4\pi \sin \theta_{\text{max}}/\lambda = 2\pi/d_{\text{min}}$, where $2\theta_{\text{max}}$ is the highest scattering angle normally used.
- (iii) ϕ_s is the neutron flux at the sample position, time-averaged and integrated over thermal energies in the TOF case. This is known with reasonable accuracy for the instruments in Table 1, but is not

necessarily optimized with respect to the source. For example, both D1A and D1B are on thermal guide tubes and are far from optimized with respect to vertical divergence, while 2XD suffers from not having a vertically focussing monochromator.

- (iv) V is the maximum "useful" volume of sample, as determined by constraints imposed by the size of the incident beam, transmission factors (assuming no highly absorbing materials), and resolution considerations.
- (v) Ω_d is the solid angle of the detector assembly, the horizontal divergence being defined by 10' Soller collimators on D1A. For the GPPD, the listed Ω_d is for the back-scattering detectors only; similarly-sized detectors are also available at other angles.
- (vi) R is the resolution of the instrument, expressed as $\Delta d/d$. For GPPD, this is approximately constant over the useful range of d spacing. For the CW instruments R varies strongly with d , and has been taken as the value at the focussing minimum, which for a high resolution machine should ideally be in the region of $2\theta = 90^\circ$, where the density of peaks is greatest. For practical comparison with TOF instruments in a typical experiment, a somewhat higher value is probably more realistic.

The product $\phi_S V \Omega_d$ is roughly proportional to the total number of neutrons detected for a complete pattern, and represents a rough figure-of-merit at that particular resolution. Thus in terms of total neutrons detected, D1A and 2XD are roughly comparable, but not in resolution, of course. This is borne out in practice by the fact that typical data sets take about the same time, T , to collect. By this criteria, the figure-of-merit for GPPD is much higher, but this is generally not found to be so in practice, since comparable data collection rates are perhaps twice that of D1A. This is attributed to the fact that the time-averaged flux at the sample contains a large proportion of short wavelength neutrons which are not necessarily useful in the subsequent data analysis for many experiments. Furthermore, since the TOF instruments exhibit a relatively constant resolution as a function of d spacing, large d spacing reflections are measured with significantly higher resolution, and therefore lower integrated intensity, than for the CW instruments. Nevertheless, when combined with a familiarity with the operating characteristics of CW and TOF diffractometers, the figure of merit, $\phi V \Omega_d$, is a useful quantitative guide to instrument performance.

Table 1: Currently-operating powder diffractometers used for performance comparisons

<u>Instrument</u>	<u>Source</u>	<u>Principal Feature</u>	<u>Type</u>	<u>Operational</u>
D1A	ILL	High resolution	CW-MC	1974
D1B	ILL	High intensity	CW-PSD	1974
GPPD	ANL	High resolution	TOF	1981
2XD	MURR	Medium resolution/ medium intensity	CW-PSD	1983

CW = Constant wavelength

TOF = Time of flight

MC = Multi-counters

PSD = Position sensitive detector

Table 2: Typical performance parameters for currently-operating neutron powder diffractometers

	<u>D1A</u>	<u>D1B</u>	<u>GPPD</u>	<u>2XD</u>
$\lambda(\text{\AA})$	1.9	2.5	-	1.3
$Q_{\max}(\text{\AA}^{-1})$	6.5	4	15	6
$\phi_s(n\text{-cm}^{-2}\text{-s}^{-1})$	2×10^6	2×10^6	$5 \times 10^5*$	6×10^5
$V(\text{cm}^3)$	3	1	5	1.5
$\Omega_d(\text{steradians})$	0.004	0.1	0.1	0.025
$R(\%)$	0.2	1.0	0.25	0.5
$\phi_s V \Omega_d$	2.4×10^4	2×10^5	$2.5 \times 10^5*$	2.25×10^4
$T(\text{hours})$	12	0.3	8	6

* Total integrated thermal flux may not be useful in all experiments.

2. New Instruments on Existing Sources

Table 3 lists the same operating parameters for new instruments which will shortly be operational or have been designed at existing neutron sources. The high-resolution and high-intensity diffractometers at ILL, D1A and D1B, will be replaced by new instruments, D2B and D20, located at the reactor face rather than on a guide tube. This will provide increased flux and allow higher resolution in the case of D2B and higher intensity in the case of D20. HRPD is a time-of-flight diffractometer on a 100 m-long guide tube at the SNS pulsed source which will soon begin operation at Rutherford Lab. For the HRPD, the flux on sample is integrated over the wavelengths propagated by the guide tube (roughly 1 - 4 Å). Thus, all of the flux will be used for most experiments and the product $\phi_s V \Omega_d$ should not need to be degraded for unused flux as was discussed for the GPPD. PADI is a high intensity and diffuse scattering diffractometer which has been proposed for the SNS. Scintillator detectors are arranged in four groups to give a total detector solid angle of 1.52 steradians. The largest detector areas are for conical arrays in forward-(7°-25°) and backscattering (138°-150°). In Table 3; only the area of the backscattering detector (138°-150°) is used for comparison with other instruments. The 2XC design is a proposed upgrade of the MURR diffractometer which benefits primarily from a more efficient, focussing monochromator and a second position sensitive detector. These instruments illustrate that diffractometer design could be pushed further at most existing sources.

Table 3: Performance parameters for advanced diffractometers at existing sources

	<u>HRPD</u>	<u>PADI</u>	<u>D2B</u>	<u>D20</u>	<u>2XC</u>
$\lambda(\text{Å})$	-	-	1.6	2.5	1.5
$Q_{\max}(\text{Å}^{-1})$	8	30	7.5	4	6.5
$\phi_s(n\text{-cm}^{-2}\text{-s}^{-1})$	10^6	5×10^7	10^7	10^8	4×10^6
$V(\text{cm}^3)$	2	5	5	0.5	1.5
$\Omega_d(\text{steradians})$	0.1*	0.77*	0.008	0.2	0.05
$R(\%)$	0.05*	0.5*	0.05	0.5	0.15
$\phi_s V \Omega_d$	$2 \times 10^5*$	$2 \times 10^8*$	4×10^5	10^7	3×10^5

* These numbers apply to the back-scattering detector array only.

3. Future Instruments on High Flux Sources

Table 4 lists operating parameters for instrument designs based on a 10^{16} n-cm⁻²-s⁻¹ reactor or a 10^{17} n-cm⁻²-s⁻¹ pulsed source. These fluxes are taken as being a factor of 10 above the ILL, a factor of 10 above SNS or a factor of 250 above IPNS. Thus, new instrument designs can be readily compared with existing instruments. As in the previous tables, the resolution, R , is nominally constant for the TOF instruments, while the R 's given for the CW instruments are the minimum values. In general, the TOF instruments exhibit a lower flux on the sample because of the $1/\lambda^2$ decrease as the incident path length, λ , is increased to achieve higher time resolution. Furthermore, the TOF instruments do not benefit from the large vertical divergence of the incident beam which can be utilized on the CW instruments with large, focused monochromators. For most TOF instruments which have the detectors in a plane, the vertical divergence of the incident beam could be increased by a large factor without degrading resolution. Unfortunately, moderators of the desired shape have not yet been developed at pulsed sources. However, the TOF diffractometers do employ the large detector areas which are possible in back-scattering. At smaller scattering angles, both designs face the same detector angular divergence requirements. The constant-wavelength, multi-counter (CW-MC) design is based on 3' soller collimators in the incident and scattered beams. The TOF designs are based on the 100 K liquid CH₄ moderator (moderator B) described by Crawford, Russell and Carpenter. This choice simplifies calculation of pulse widths and fluxes. Guide tubes used in the TOF designs are assumed to be nickel [$\theta_c = (0.11 \lambda)^\circ$, for λ in Å] with 50% transmission efficiency at 100 m. The application of supermirrors could increase the guide tube gain by as much as a factor of 2, assuming that they could be used only to increase the vertical divergence. (For guide tube designs, the horizontal divergence is already optimized.) A brief description of each instrument design follows.

Table 4: High-resolution and high-intensity diffractometers for future, high-flux sources -- 10^{16} n-cm⁻²-s⁻¹ reactor or 10^{17} n-cm⁻²-s⁻¹ pulsed source

	CW-PSD	CW-MC	TOF-7m	TOF-65m	TOF-130m
ϕ_s (n-cm ⁻² -s ⁻¹)	10^9	5×10^7	9×10^8	$4 \times 10^6^*$	8×10^6
V (cm ³)	2	5	1	4	1
Ω_d (sterrad.)	0.15	0.005	0.2	0.1	0.1
R (%)	0.5	0.03	0.5	0.06	0.03
$\phi_s V \Omega_d$	3×10^8	1×10^6	2×10^8	$2 \times 10^6^*$	8×10^5

* assuming no guide tube is used.

CW-PSD: Constant-Wavelength, High-Intensity Diffractometer

Present powder diffraction instruments, such as the D1B instrument at ILL or the 2XD-PSD instrument at the University of Missouri are designed for rapid data collection (1-6 hours for a full diagram, 1-10 minutes for rapid survey) and sacrifice resolution ($\Delta d/d \approx 1\%$ and 0.5% respectively) for that purpose. Even these machines are far from optimum for the present generation of reactor sources and more than an order of magnitude gain should be realized by improved beam transport, focusing, high-reflectivity monochromators and larger detectors. A high efficiency instrument, for experiments such as time resolved studies would utilize the same design principles as those presently in use at the state-of-the-art instruments-- a high flux on the sample and a large product of detector solid angle and sample size. The principles of beam transport are well known and need no major elaboration. It is quite clear, however, that for maximum efficiency one needs large source area (especially height), short source to sample distance and a focusing monochromator. At an optimized 10^{16} reactor, it is reasonable to expect $\sim 10^9$ n-cm⁻²-s⁻¹ on the sample. Two major options exist for the sample-detector combination, multiscanner systems with Soller slit collimators and position sensitive detectors (PSD). Since the resolution of the PSD system depends upon the sample diameter as well as the diffraction optics, the figure of merit for a sample-detector combination depends on the product of solid angle and sample size and, hence, cannot be increased indefinitely with either method, due to the space limitations for the Soller slit system or the sample size limitation for the PSD. Since the optimized product is roughly equal for the two schemes, it is sufficient to describe only one.

A representative instrument can be scaled from existing designs. Constructed from presently available components (24" x 1" linear position sensitive 10 Atm ³He detectors), an ensemble of 30 elements would cover roughly 120° horizontal x 4° vertical, at 1.3Å would be $\sim 80\%$ efficient, and would have $\sim 0.5\%$ resolution at the minimum, ($2\theta \approx 2\theta_m$). With a 1 cm diameter, 10% scattering sample the count rate would be $\sim 3 \times 10^7$ /sec over the entire detector, a figure which exceeds presently operating instruments by about three orders of magnitude.

The applications of such an instrument are quite exciting. With throughputs of this magnitude, strong reflections could be seen in $\sim 10^{-3}$ sec, and time resolved studies could be performed on a time scale sufficiently short to study reaction kinetics, phase transitions, etc. The ease with which special environments could be provided for neutron scattering, combined with this speed may provide a significant advantage over comparable studies with other probes. One

particularly attractive advantage may be the ability to observe and refine the structures of short-lived intermediate products which are produced in a chemical reactor mounted on the neutron diffractometer.

The most challenging aspect of such a device is the need for electronics capable of processing $\sim 10^6$ events/s per detector. While progress has been made in detector speed in the last decade, this represents a quantum step from present technology which should be pursued vigorously.

CW-MC: A High Resolution Powder Diffractometer on a 10^{16} Reactor

The design for such an instrument follows the principles employed for D2B at ILL Grenoble (Hewat 1975) but provides higher resolution and higher intensity. These gains will be important for the solution of structures from neutron powder data by direct methods, and for the refinement of structures more complex than now possible.

The new machine and D2B are compared in Tables 1 and 4. The important parameters are the useful flux on the sample, the sample volume, the solid angle subtended by the multidetector, and of course, the resolution. The product of flux on the sample, sample volume and detector solid angle is proportional to the total number of neutrons collected. The quoted resolution is the value at the minimum, which should probably be increased somewhat for comparison with TOF instruments (which have nominally constant resolution).

The resolution design goal is $\Delta d/d = 3 \times 10^{-4}$ using 3' soller collimators before the monochromator and before each of 64 counters. Such collimators are a feasible extrapolation of the existing 5' D2B collimators.

The flux on the sample may be obtained using the formula given by Moon

$$I(\lambda) = \frac{\phi_0}{4\pi} \gamma_\lambda(T_0\lambda) P_M(\lambda) \delta_V \lambda \cot \theta_M \left(\frac{2\pi}{8\lambda n^2} \right) \frac{\alpha_0 \alpha_1 \eta_M}{(\alpha_0^2 + \alpha_1^2 + 4\eta_M^2)^{\frac{1}{2}}}$$

$\phi_0 = 1 \times 10^{16}$ is the reactor flux, $\gamma_\lambda = 0.7$ its wavelength distribution function at $\lambda = 1.6 \text{ \AA}$, $\delta_V = h_M/L_1$ the ratio of the height (300 mm) of the vertically focussing monochromator to its distance (2500 mm) from the sample, $\theta_M = 77.5^\circ$ the monochromator angle, $\alpha_0 = 3'$ and $\alpha_1 = 30'$ the collimation before and after the monochromator, and $\eta_M = 15'$ the monochromator mosaic spread. $P_M(\lambda)$, the crystal reflectivity, is taken as 0.5. Then

$$I(\lambda) = \left(\frac{1 \times 10^{16}}{4\pi} \right) (0.7)(0.5)(0.12)(1.6)(0.4)(1.13) \frac{\times 3 \times 30 \times 15}{(9 + 900 + 1600)^{\frac{1}{2}}} \times \left(\frac{1}{60} \right)^2 \times \left(\frac{\pi}{180} \right)^2.$$

$$I(\lambda) = 5 \times 10^7 \text{ n-cm}^{-2}\text{-s}^{-1}$$

Thus, the new high flux reactor machine would be roughly four times as fast as D2B at ILL ($\phi_s V \Omega_d = 1.8 \times 10^6$ compared to 4×10^5 on D2B) while having considerably better peak resolution ($\Delta d/d = 3 \times 10^{-4}$ compared to 5×10^{-3} on D2B). One possible improvement would be to double the detector height, doubling Ω_d and doubling the number of useful neutrons detected at the expense of larger peak asymmetry corrections.

TOF-7 m: High-Intensity, Time-of-Flight Powder Diffractometer

The performance of a high intensity, backscattering TOF diffractometer on a 10^{17} pulsed source can be estimated by multiplying the flux by a factor of ten for PADI (Table 3). However, many useful high intensity experiments, particularly in special environments, cannot be done in backscattering. Thus, it is useful to consider a design optimized at 90° .

For the high-intensity, time-of-flight diffractometer included in Table 4 the design goal is resolution ($\Delta d/d$) of 0.005 at a scattering angle of $2\theta = 90^\circ$, which would permit optimal collimation for use of the instrument for special environment diffraction. For moderator B, the pulse width Δt is 9 μs at 50 meV (1.28 Å). For a total flight path of length λ , this is equivalent to

$$\frac{\Delta t}{t} = \frac{0.0278}{\lambda}$$

where λ is in meters. If the time and geometrical resolutions are made equal

$$\frac{\Delta t}{t} = \frac{0.005}{\sqrt{2}} = .0035$$

which gives

$$\lambda = \frac{.0278}{.0035} = 7.94 \text{ m.}$$

Assuming the incident and scattered angular divergences are equal,

$$\Delta \theta \cot \theta = \frac{.0035}{\sqrt{2}} = .0025$$

so that at $2\theta = 90^\circ$

$$\Delta\theta = \frac{0.0025}{\cot 45^\circ} = 0.0025$$

but

$$2\Delta\theta = \frac{w}{\lambda_1} = \frac{D}{\lambda_2}$$

where w is the moderator width, D is the detector diameter (assuming cylindrical ^3He proportional counters) and λ_1 and λ_2 are the incident and scattered flight paths. Since the instrument will be most heavily used for small samples, we choose $D = 0.5$ cm which gives

$$\lambda_2 = \frac{0.5}{.005} = 100 \text{ cm} = 1 \text{ m}$$

Then, since $\lambda_1 + \lambda_2 = 8 \text{ m}$, we have $\lambda_1 = 7 \text{ m}$, and $w = 0.005 \times 7 = .035 \text{ m}$.

For best performance, the moderator for this instrument should be 3.5 cm wide and 20 cm high. Such a moderator could probably be coupled to the target with efficiency similar to a 10 x 10 cm moderator. As an alternative, a 10 x 10 cm moderator could be used with a mask near the source, or a Soller collimator near the sample, to reduce the effective width to 3.5 cm for high resolution (0.5 %) measurements. For lower resolution measurements, the entire moderator could be viewed and larger, 1 cm diameter, samples could be employed giving roughly an order of magnitude increase in count rate and resolution of 0.9 %.

The flux on the sample is given by (following Crawford, Russell and Carpenter)

$$\phi_s = \frac{I_{th}}{\lambda_1^2}$$

Using the values for moderator B on a $10^{17} \text{ n-cm}^{-2}\text{-s}^{-1}$ pulsed source, we get

$$\phi_s = \frac{\bar{\phi}_{eff} A}{4\pi \times 700^2} = \frac{7.9 \times 10^{13} \times 3.5 \times 20}{4\pi \times 700^2} = 9.0 \times 10^8 \text{ n-cm}^{-2}\text{-s}^{-1}$$

where the moderator area A is taken as $3.5 \times 20 \text{ cm}^2$. The sample is assumed to be 0.5 cm diam by 5 cm high which gives a sample volume of 1 cm^3 . Larger samples, up to 5 cm^3 , could readily be accommodated with a loss of resolution as previously discussed or with $\lambda_2 = 2 \text{ m}$ rather than 1 m . Experience with existing instruments shows that a detector solid angle (time-focused) of 0.2 steradians could easily be achieved for this design.

TOF-65 m: High-Resolution Time-of-Flight Powder Diffractometer

Since the TOF diffractometers exhibit constant resolution over a wide range of d -spacings, a reasonable design goal for comparison with constant-wavelength instruments is about twice the minimum CW resolution. Thus, the design goal (for comparison with CW-MC) is $\Delta d/d = 6 \times 10^{-4}$.

Assuming, time and geometrical resolutions are equal, (for moderator B)

$$\frac{\Delta t}{t} = \frac{6 \times 10^{-4}}{\sqrt{2}} = \frac{0.0278}{\lambda},$$

which gives

$$\lambda = 65.5 \text{ m}.$$

Assuming a moderator 10 cm wide, the incident divergence (which is set equal to the scattered divergence) is matched at a scattering angle of

$$\cot \theta = \frac{6 \times 10^{-4}}{2\Delta\theta} = \frac{6 \times 10^{-4}}{2 \left(\frac{\omega}{2\lambda} \right)} = \frac{6 \times 10^{-4}}{2 \left(\frac{.10}{2 \times 65} \right)}$$

$$\theta = 68.7^\circ.$$

By going further into back-scattering ($2\theta > 137^\circ$), large detector solid angles can be achieved. At small scattering angles, the detector angular divergence requirements are nominally equivalent to those for a CW instrument except that longer wavelength neutrons can be used to advantage in the TOF case on a cold (100K) moderator. Choosing $2\theta = 160^\circ$, the incident divergence becomes negligible and for a 1 cm diameter detector we get a scattered flight path of

$$\lambda_2 = \frac{1}{2 \times \frac{6 \times 10^{-4}}{\sqrt{2}}} \cot 80^\circ = 207 \text{ cm}.$$

At this detector distance, it is possible to readily achieve a time-focused detector solid angle of 0.1 steradians. The sample can be 1 cm diam and 5 cm high; $V = 4 \text{ cm}^3$.

The flux on the sample is given by

$$\phi_s = \frac{1}{4} \frac{I_{th}}{l^2} = \frac{1}{4} \frac{\bar{\phi}_{eff} A}{4\pi \lambda_1^2} = \frac{1}{4} \frac{7.9 \times 10^{13} \times 100}{4\pi \times 6500^2} = 3.7 \times 10^6 \text{ n cm}^{-2} \text{ s}^{-1}$$

where the factor of 1/4 is for the reduction in repetition rate (from 50 hz to 12.5 hz) required at this path length to avoid frame overlap.

A two-sided guide tube could be used to increase the flux by increasing (only) the vertical divergence. The possible gain for a nickel guide (assuming 75 % transmission efficiency) is given by

$$\frac{0.11 \lambda}{\left(\frac{.1}{2 \times 65} \times \frac{180}{\pi}\right)} = \times .75 = 1.87 \lambda$$

where λ is in Å. For 2 Å neutrons this gives a gain of 3.7 and the approximate flux on sample would be

$$\phi_s = 1.4 \times 10^7 \text{ n cm}^{-2} \text{ s}^{-1}$$

It should be noted that for the cold moderator used in this design, more neutrons of useful wavelengths are included in the time-averaged, integrated thermal flux, ϕ_s , and one need not downgrade ϕ_s as was previously discussed for existing TOF instruments when making comparisons with CW instruments.

TOF-130 m: High-Resolution Time-of-Flight Powder Diffractometer

It is useful to make an estimate of the performance of a diffractometer with resolution of $\Delta d/d = 3 \times 10^{-4}$. Such a machine can be simply scaled from the HRPD at SNS. The required time resolution, $\Delta t/t$, is $3 \times 10^{-4}/\sqrt{2}$, which, for moderator B, requires a path length of

$$\lambda = \frac{0.0278}{(\Delta t/t)} = \frac{0.0278 \times \sqrt{2}}{3 \times 10^{-4}} = 131 \text{ m}.$$

If a curved guide tube is used on a cold moderator, we can estimate the flux on sample by scaling the SNS flux ($10^6 \text{ n-cm}^{-2}\text{-s}^{-1}$) by a factor of ten and assuming an additional guide tube loss of (100/131). Thus,

$$\phi_s = 10^6 \times 10 \times \frac{100}{131} = 8 \times 10^6$$

It should be noted that even though the guide tube allows a sizeable gain in flux (e.g., as compared to the 65 m - TOF instrument), the incident angular divergence is increased significantly, which degrades the resolution at all angles except backscattering. The repetition rate and the detector configuration ($\Omega_d = 0.1$ steradians) could be about the same. When the detector covers the entire cone in backscattering, the possible sample volume is reduced; $V = 1 \text{ cm}^3$. Thus, the overall performance is given by

$$\phi_s V \Omega_d = 8 \times 10^5.$$

Since the guide passes only a band of wavelengths in the useful thermal range, this flux should not need to be degraded for comparison with CW instruments.

Comparison of Powder Diffractometers on Reactors and Pulsed Sources and Conclusions

Based on current experience and projected instrument performance for both reactors and pulsed neutron sources, some fairly clear conclusions can be drawn about the advantages or disadvantages of each technique for particular kinds of measurements. These conclusions are summarized in Table 5.

For the case of structural refinement, which is expected to continue to be the major use of the powder technique, neither type of instrument holds a clear advantage. Present TOF designs suffer from lower fluxes on the sample since long flight paths are required to achieve high resolution, but they can take full advantage of the large detector areas which can be employed in back-scattering. While current CW diffractometer designs are quite advanced, TOF designs have not yet fully exploited the gains which could be realized through the use of rectangular moderators (to increase vertical divergence), guide tubes combined with Soller collimators and super-mirror guides. Moreover, most present-day TOF diffractometers do not take full advantage of the data collected at angles other than back-scattering. By collecting data at multiple, fixed angles, a more restricted range of wavelengths could be used and the repetition rate (and flux on sample) could be increased a factor of 2-4, in addition to the gain in "active" detector area.

For indexing phases and structure solution, which require deconvolution of the raw data into individual peak positions and intensities, both types of instruments are expected to perform well as resolution is improved. However, for

CW instruments, the provision for long wavelengths is needed and, for TOF instruments, small scattering angles must be provided (with suitably high resolution) to accurately measure the reflections with long d-spacings.

The pulsed source instruments will continue to provide a more precise refinement for those cases where the data analysis can be extended to high Q (small d-spacing). Such systems are typically hard lattices with small or moderately-sized unit cells.

For magnetic structures, both types of instruments are capable of observing the small q magnetic reflections. However, in some cases the CW instruments may offer the operational advantage of measuring both nuclear and magnetic reflections in the same scan which is useful for estimating absolute magnetic intensities. In the TOF case at low fixed angles, the scaling of magnetic to nuclear intensities may be more difficult. However, additional experience in the TOF case may resolve this difficulty.

For high data rate diffraction, both types of instruments will perform well. If the experiment is best done at a fixed scattering angle, the TOF method offers an important advantage. This advantage could be exploited for the study of a localized region of a bulk sample. The best high pressure diffraction will also be done at a fixed scattering angle (usually 90°) where collimation can be most effectively used to eliminate cell scattering. The same arguments apply to those extreme temperature sample environments where sample containers or heat shields must be placed very close to the sample (making collimation difficult except at $2\theta = 90^\circ$).

Table 5. Relative advantages of the projected 10^{16} reactor or 10^{17} pulsed source powder diffractometers for various types of measurements. Key: x = no clear advantage, 0 = disadvantage, yes = clear advantage.

	<u>10^{16} Reactor</u>	<u>10^{17} Pulsed Source</u>
Refinement	x	x
Solution	x (long λ)	x (low θ)
High-Q	0	yes
Magnetic	maybe	?
Time-dependent	x	x
Small-sample	x	x
High pressure	0	yes

Crystallography (Single Crystal and Protein)

T. F. Koetzle, Brookhaven, Co-Chairman
B. P. Schoenborn, Brookhaven, Co-Chairman
U. W. Arndt, Cambridge
J. K. Blasie, Pennsylvania
R. A. Jacobson, Iowa State
M. S. Lehmann, Grenoble
S. A. Mason, Grenoble
A. J. Schultz, Argonne

Recommendations

Estimates comparing both medium and high-resolution single-crystal diffractometers on the proposed advanced steady-state reactor and pulsed spallation sources indicate that approximately equivalent performance could be expected for the two types of advanced sources (vide infra), with gains of 10-100 over present instruments. However, additional effort will be required at the pulsed source to make corrections for wavelength-dependent extinction, multiple-beam scattering, and thermal diffuse scattering. Two-dimensional position-sensitive detectors will be used for all single-crystal diffractometers, and the higher fluxes will introduce stringent counting-rate requirements. For an advanced steady-state reactor, existing state-of-the art devices should be adequate, but for a pulsed source some development will be necessary to cope with higher instantaneous counting rates. It is recommended that support for development of improved high-resolution position-sensitive detectors be given immediate high priority; for maximum efficiency, such an effort should involve close collaboration among existing U.S. neutron scattering centers. Supermirror guides will be required for high-resolution diffractometers at an advanced pulsed source, to transmit neutrons efficiently over the long flight path (> 20 m), and it is recommended that

a pilot research program be undertaken now to solve the technical problems associated with the manufacture of these guides.

In general, instrument design should be flexible to allow new options and facilities to be implemented as new scientific needs emerge. A wide range of sample environments must be possible, for example, low temperatures and high pressures for the study of superconductors, for studies of proteins under pressure and for cryoenzymology. In all applications, optimal data extraction requires computer analysis of the entire scattering pattern recorded by position-sensitive detectors.

The application of pulsed sources to single-crystal neutron diffraction is a development of the past few years. In order to allow an experimental comparison of existing instruments at pulsed and steady-state sources, benchmark data on a standard sample should be collected at as many centers as feasible, in the near future.

Scientific Justification

Single-crystal neutron diffraction provides a uniquely sensitive technique for structural studies. A number of characteristics of the neutron scattering process are exploited in neutron crystallography.

1. Neutrons interact with the atomic nuclei in the sample, and thus the information about atomic positions and thermal vibration parameters is not biased by electron density distributions.
2. Atomic scattering cross sections vary more-or-less randomly across the periodic table, enabling neighboring elements and different isotopes of the same element to be distinguished readily, in most cases.
3. The neutron possesses a magnetic moment, and diffraction with polarized neutron beams yields information on magnetic structures and electronic spin-density distributions.
4. Neutrons have great penetrating power and are therefore very useful for studies of samples in special environments, e.g., extreme temperatures and pressures, magnetic fields, etc., and for diffraction from bulk samples.
5. Slow neutrons do not cause sample radiation damage, and it is possible to carry out a complete diffraction experiment with one crystal.

In many applications, the information obtained from neutron crystallography is complementary to that obtained from x-ray experiments.

Single-crystal neutron diffraction is an essentially signal-limited technique. Many high-precision studies on complex chemical systems require from several weeks to a month of beam time on conventional single-counter instruments. The use of two-dimensional position-sensitive detectors, pioneered for protein crystallography, is just beginning for materials with smaller unit cells ($d \lesssim 25$ Å) but will cut this time to a few days. The order-of-magnitude increase in flux expected from the next generation of sources will make possible a number of new types of experiments, for example, by improving the Q resolution to resolve larger unit cells, making it possible to use smaller samples and to carry out series of investigations as a function of temperature and pressure or on related compounds within a reasonable period of time, and to undertake time-dependent studies. The importance of being able to carry out experiments with smaller samples should be emphasized, as the range of materials amenable to examination by single-crystal neutron diffraction presently is limited severely by the requirement for crystals of size $> 1 \text{ mm}^3$.

A traditional area of neutron crystallography has been the study of hydrogenous materials, such as metal hydrides, organometallic compounds, and hydrogen-bonded systems. These continue to be areas of great interest. For example, metal hydride structures are relevant to hydrogen storage applications as well as hydrogenation and dehydrogenation reactions, while organometallic systems exhibiting C-H...metal interactions are models for C-H bond activation, central to many important processes such as Fischer-Tropsch synthesis and olefin metathesis. Achievement of the flux goals set forth for the advanced sources together with development of improved position-sensitive detectors would make it possible to study crystals of size approaching that used in conventional x-ray crystallography, and to greatly expand the application of neutron diffraction to chemical problems. New applications may develop, e.g., the rapid determination of hydrogen and deuterium distributions to probe hydrogen exchange and absolute stereochemistry.

In recent years, neutron diffraction has been applied to framework structures such as zeolites. These experiments are giving the first reliable information on the orientation and hydrogen bonding properties of water molecules in the zeolite channels, and temperature-dependent studies are yielding important insight into the process of dehydration in these molecular sieves. It has been possible to discern details of Al/Si ordering in the zeolite frameworks which cannot be deduced from x-ray results. The catalytic properties of zeolites are determined by the interaction between substrates, solvent molecules and the framework, so that these experiments are highly relevant to industrial applications. The neutron diffraction work is, however, severely limited at present because the great majority of zeolites do not form single crystals of adequate size.

The unbiased atomic positions and thermal vibration parameters from neutron diffraction studies can be combined with x-ray diffraction to provide information on electron-density distributions in solids. These investigations, begun in the 1960's for organic compounds, have more recently been extended to systems containing heavier elements, including transition-metal compounds. The availability of increased neutron flux will make it possible to reduce the sample size, thus minimizing systematic errors due to extinction and leading to more precise results. It may also be possible to carry out the neutron and x-ray measurements on the same crystal, which would be highly desirable. Charge-density studies require data to very high Q , and short-wavelength neutrons from a spallation source or a hot moderator at a steady-state reactor will make it possible to obtain intensity data in the desired range.

The distributions of unpaired p and d electrons in paramagnetic samples oriented in a magnetic field can be probed with polarized neutron beams. The results of both charge and spin-density investigations can be compared with quantum mechanical calculations, to yield important insights into the nature of chemical bonding. The spin-density measurements have so far been confined to quite simple systems, because the measurements are extremely time consuming, so that it is not possible to obtain magnetic structure factors for more than a few hundred reflections to the required precision of a few percent relative error.

Studies of ionic conductors exploit the fact that the nucleus is an effective point scatterer, so that the atomic scattering cross-section is constant with Q and correlation between site occupancy and thermal motion is minimized. Recent experiments have involved the investigation of materials such as γ -alumina, AgI, Ag₂S, Cu₂S and related ternary systems as a function of temperature, and yttrium stabilized zirconium oxide under an applied electric field. Because of the delocalized distributions of mobile ions, diffracted intensities from ionic conductors usually are weak, and increased flux will lead to increased sensitivity to details of the distributions.

Use of the Laue technique with a two-dimensional position sensitive detector or film provides an extremely attractive method for materials examination, e.g., topographic measurements on bulk samples, and examination of extended portions of the reciprocal lattice for super-lattice peaks and diffuse scattering. This capability has been extremely important in work on incommensurate structures such as the organic superconductors.

Pulsed neutron sources provide an excellent opportunity to study wavelength dependence of extinction and obtain a better understanding of this effect and how to correct for it. As the crystal is rotated, individual reflections move across the face of a two-dimensional detector and the wavelength for each reflection varies continuously. This then affords an excellent means to obtain intensity vs. wavelength information on a large number of reflections, for which the numerous theoretical approaches to account for extinction can be applied and evaluated. In general, thermal diffuse scattering also varies with wavelength, and it is imperative to make adequate corrections for this effect.

Time-dependent applications of neutron crystallography may become possible at the advanced sources. If the time required to obtain a complete set of Bragg intensity data can be reduced to on the order of hours or minutes, kinetics of slow solid state chemical reactions may be studied in real time. Polymerization induced by light or pressure could be followed in single-crystal samples, as could the kinetics of melting and intercalation processes. A pulsed source would be particularly useful for studying reversible processes stimulated by an external probe that

could be timed with respect to the incident neutron pulse. For example, the response of dielectric materials to a varying electric field could be explored in this way.

In neutron protein crystallography, serious interpretation of the significant new information from near-atomic resolution has only become possible in the last 10 years. The most important results are derived from the unique ability to measure the degree of H/D exchange of almost all the labile protons in a protein, for much higher molecular weight systems than is possible with solution n.m.r. This has led to a new understanding of the dynamics of protein molecules and their flexibility, and has given insights into the functioning and into the denaturation and folding of proteins. However, in order to obtain kinetic information, each stage of the experiment must be completed as rapidly as possible, preferably in a few days, in order to limit changes in the sample while any one stage is measured.

Recently, at least three new areas of fundamental interest have emerged:

1. Solvent regions between molecules, salt bridges, etc., can be examined in great detail. Neutron diffraction is a powerful technique here because proteins can be studied in both D_2O and H_2O (or even mixtures) and, employing sophisticated phasing techniques, new information can be extracted even for partially-ordered solvent.

2. Distortions of protein geometry from ideality can be studied, but only if data can be measured to atomic resolution. This requires recording at least 50,000 reflections even for a medium-sized protein such as lysozyme (MW 14000). Here neutrons are the only technique with the power to define geometry where H atoms are involved, e.g., hydrogen-bonding parameters. Note that while neutron diffraction gives roughly twice as much information as the corresponding x-ray experiment, since H atoms are located, twice as many neutron intensity data also are required.

3. Experiments to investigate the nature of the interactions between proteins and surface probes such as deuterated alcohols are of general interest to protein chemists. Such species can be seen at very high contrast relative to non-deuterated solvents, making it possible to study the localization of these molecules on protein surfaces.

For completeness, it should be noted that contrast variation techniques may also be applied readily to single crystals with quite large unit cells -- several hundred Angstroms on edge -- at present to limited resolution (d spacings ~ 10 Å). Thus the spatial organization of different structural components (i.e., protein, nucleic acid, ...) can be inferred. Here, there is a critical shortage of beam time which prevents the technique from becoming generally useful: In fact, there is only one instrument in existence designed expressly for this purpose.

In each of the areas of protein crystallography currently seen to be important, a gain in "overall experimental efficiency" of a factor of 10-100 (e.g., a factor of 10 from larger position-sensitive detectors and 10 from source flux and better optics) is needed in order to extend the range of macromolecules that can be studied. These would include larger proteins, or proteins which crystallize in unfavorable space groups, or which yield single crystals smaller than the present minimum acceptable volume of ~ 1 mm³. Further, sequential high-resolution structure determinations along an enzyme reaction or H/D exchange pathway would then no longer be exceptional.

The elastic scattering of neutrons from partially-ordered macromolecular systems, including the more general classes of smectic and nematic liquid crystals, can provide valuable structural information concerning the time-averaged intramolecular configurations and intermolecular ordering in these systems. The neutron scattering technique is most powerful when coupled with variation of the scattering contrast of a particular molecular species or sub-molecular fragment (including a single atomic site) in such systems, as can be achieved with selective deuteration. The level of detail in terms of spatial resolution and/or positional accuracy in such structural studies depends upon both the precision of the intensity measurements and the observability of generally low-intensity reflections at high Q . Hence, the experiments depend critically upon the incident neutron flux, and factors in excess of a ten-fold improvement in incident flux would dramatically enhance capabilities in this area. For example, a hundred-fold increase in incident flux would permit the spatial resolution achievable in the profile structures of oriented multilayers, such as membrane-type systems, to be improved from the current 15-25 Å range to 3-5 Å.

Instrumental Comparison

In this section we shall compare the diffraction experiment as carried out at the two kinds of sources. We shall assume the use of an area detector, and we shall in a first step consider instruments with identical collimations, i.e., sample to source distances are comparable and monochromator effects are neglected. Under these conditions it seems intuitively obvious that a comparison must be based on the time-averaged flux integrated over the useful wavelength range, but we shall use a general argument to support this. For the following discussion we then consider perturbations caused by effects peculiar to each source, and finally we give some flux estimates.

General Considerations

The spectral width, $\Delta\lambda$, is given by the system (unit cell and desired resolution). This leads to an illuminated volume ΔV (shaded part):

and an integrated, time averaged flux

$$\hat{\phi}_{st} = \int_{\Delta\lambda_{st}} \phi_{st}(\lambda) d\lambda, \quad \hat{\phi}_{pu} = \frac{1}{T} \int_{\Delta\lambda_{pu,T}} \phi_{pu}(\lambda, t) d\lambda dt$$

where st = steady-state and pu = pulsed source. T is a time long compared to the repetition rate. The flux per reciprocal volume unit is

$$\bar{\phi} = \hat{\phi} / \Delta V$$

and the number of illuminated lattice points is

$$n = \Delta V .$$

(We shall assume units so that all proportionality factors are unity, and the limiting case where the density of lattice points approaches the resolution limit.)

The total scattered Bragg intensity is

$$I = \overline{\phi} \cdot n .$$

Then the intensity per reflection is

$$I/n = \overline{\phi} = \hat{\phi}/\Delta V \text{ (neutrons/sec)}$$

and the time per neutron detected is

$$t = n/I = \Delta V/\hat{\phi} \text{ (sec/neutron)} .$$

For every 'frame' of the pulsed source (often called a histogram) we need N 'frames' for the steady-state source

where $N = n_{pu}/n_{st} = \Delta V_{pu}/\Delta V_{st}$.

For the pulsed source n_{pu} reflections are recorded simultaneously, and the time to detect one neutron for each is

$$T_{pu} = \Delta V_{pu}/\hat{\phi}_{pu} .$$

For the steady-state source the time for recording the same n_{pu} reflections with one neutron for each is

$$\begin{aligned} T_{st} &= N \cdot \Delta V_{st}/\hat{\phi}_{st} \\ &= \Delta V_{pu}/\hat{\phi}_{st} . \end{aligned}$$

The gain factor $G = T_{st}/T_{pu}$ is thus

$$G = \hat{\phi}_{pu} / \hat{\phi}_{st} .$$

Discussion

For the advanced sources the integrated, time-averaged fluxes on the sample are estimated to be in the neighborhood of 10^8 n/cm² · sec, if we integrate over the wavelength ranges appropriate for the two kinds of experiment. To a first approximation the factor G is therefore near unity. For two different instruments covering medium and large unit-cell size some details are given in Table 1 (pulsed source) and Table 2 (steady-state source).

We can then discuss in more detail our various assumptions:

1. We have assumed that $\phi_{st}(\lambda)$ and $\phi_{pu}(\lambda, t)$ are constant over the range of the integration in λ . Moreover we did not consider reflectivity. However, this would influence the distribution of counts as a function of momentum transfer, Q , and the counting statistics.

It would therefore be more appropriate to operate with a beam-distribution and reflectivity-weighted flux for the pulsed source. For the steady-state source we can retain an unweighted value due to the small $\Delta\lambda$.

The number of lattice points for a given d spacing is proportional to d^2 which for a limited range of scattering angle (e.g., a smallish area detector) is proportional to $k^2 = (1/\lambda)^2$. Therefore

$$\phi^*(\lambda, t) d\lambda = k^2 \phi(\lambda, t) dk = \phi(\lambda, t) / \lambda^4 d\lambda .$$

However, the reflectivity is proportional to λ^2 , and we shall include this to get $\phi^{**}(\lambda, t) = \phi^*(\lambda, t) \lambda^2 = \phi(\lambda, t) / \lambda^2$. So our weighted integrated, time-averaged flux becomes

$$\hat{\phi}_{pu}^{**} = \frac{1}{T} \int_{\Delta\lambda, T} \phi_{pu}(\lambda, t) / \lambda^2 d\lambda dt / \int_{\Delta\lambda} d\lambda / \lambda^2$$

The gain factor for this is $G^* = \hat{\phi}_{pu}^{**} / \hat{\phi}_{pu}$. A simple numeric estimate gives for $\lambda_{min} = 1.0$ Å and $\lambda_{max} = 2.0$ Å

$$G^* = 5$$

2. We only detect a limited part of the illuminated region of reciprocal space. It is therefore necessary to ensure that the proportionality between the limited region and the total illuminated volume is independent of $\Delta\lambda$. In the figure below a realistic case is illustrated, where the detector covers the range from 0 to 120° in 2θ . The shaded area is the part viewed in the pulsed-source case, and the dark area is that viewed for the steady-state source. The three-dimensional shape of the detector is assumed to be a spherical cap with ∞ -symmetry

along the incoming beam. (In practice this would be made up of a number of flat position - sensitive detectors.) Inspection shows that the assumption is reasonable.

3. We have assumed that the pulsed-source volume could be surveyed by N positions of the steady-state illuminated volume. This assumption can be studied in the above Figure. For a $2\theta_{\max}$ of 120° this is not an unrealistic assumption, and for smaller $2\theta_{\max}$ the approximation gets better. It is less obvious, though, that the two methods are equally efficient in scanning reciprocal space. For the steady-state instrument with a limited wavelength range many measurements have been made and optimized strategies have been developed. This still remains to be done for pulsed sources, and could eventually reveal unforeseen problems. Data collection strategy is discussed in more detail below under Detector Requirements.

Instrumentation

Representative instrument parameter values for instruments at 10 m and 20 m from a source at (i) a pulsed source and (ii) a steady-state source are given in Table 1 and 2, respectively. A 10 x 10 cm source size is assumed in each case; for the pulsed source the flux given is weighted as described above.

Conclusion

From the above, our estimate of instrumental "gain factors" comparing the proposed advanced steady-state to proposed pulsed source are near unity for both the medium and high-resolution instruments. In the case of the high-resolution instrument for moderate-size unit cells (approximately 25 Å on edge) the pulsed source seems to have a higher flux performance than the steady state. However, as the pulsed-source instrument is based on a multi-wavelength method, a series of diffraction effects might well become of importance in limiting the accuracy of the observed structure factors, notably extinction and the related multiple-beam scattering. Even though theoretical models are available to correct for these effects, it is not usual for a crystal to follow any simple model for crystal mosaicity and defects, and the model will then only work for a limited wavelength range. The solution to the problem would most often be to reduce crystal size. The apparent gain factor should therefore be taken as the upper limit. Thus, we do not foresee any large advantage or disadvantage with either source. We have not tried to optimize all the instrument factors, but clearly we would expect a gain of 10-100 in performance over present instruments.

Table 1. Instrument Parameters for Time-of-Flight Single-Crystal
Diffractometers

	<u>I</u>	<u>II</u>
L_1 (source-to-sample)	10 m	20 m
L_2 (sample-to-detector)	1 m	
detector size	$20^\circ \times 20^\circ$	
$\Delta t/t$ (moderator A)	0.0087	0.0044
$\Delta L/L = 0.02$ m/L	0.002	0.001
crystal mosaic	0.1°	
crystal size	$2 \times 2 \times 2$ mm	
detector resolution = $20^\circ/100$ elements	0.2°	
overall $\Delta\theta$ (radians)	0.011	0.007
<u>at $2\theta = 90^\circ$</u>		
$R = \Delta S/S$ (resolution)	0.014	0.008
$(S = 2 \sin\theta/\lambda)$		
<u>for cell dimension $a =$</u>	25 Å	100 Å
$d_{\min} = R(2a)$	0.7 Å	1.6 Å
$\lambda_{\min} = 2d_{\min} \sin 45^\circ$	1.0 Å	2.3 Å
$\phi_{pu}^{**} (n/cm^2 \cdot \text{sec})$	6×10^8	3×10^7
λ range (Å)	1.0-2.0	2.3-4.0

Table 2. Instrument Parameters for Steady-State Single-Crystal
Diffractometers

Expressions used:

$$\hat{\phi}_{st}(\lambda) = \phi''(\lambda) P_M(\lambda) \delta_V \lambda \cot \theta_M \cdot 1.13 \cdot \frac{\eta_M^{-2}}{\sqrt{6}}$$

$$\Delta\lambda/\lambda = \cot \theta_M \bar{\eta}_M / \sqrt{2}$$

(reference: R. Moon: Workshop Introduction for Steady-State Reactors).

This assumes matched divergence and monochromator mosaic with monochromators in transmission geometry so that some 'horizontal focusing' is obtained. Similar effects might be obtained using reflection geometry for the monochromator and horizontal focusing, which explains $P_M > 0.5$.

	<u>I</u>	<u>II</u>
L_1 (source-monochromator)	10 m	20 m
L_2 (monochromator-sample)	2 m	
λ	1.0 Å	1.5 Å
$\phi''(\lambda)$ (n/cm ² · sec)	3×10^{14}	2.5×10^{14}
θ_M	20°	20°
$\bar{\eta}_M = \alpha_0$	10/1200	10/2200
P_M	0.5	0.7
$\hat{\phi}$ (n/cm ² · sec)	1.1×10^8	3.4×10^7
$\Delta\lambda/\lambda$ (~ $\Delta\theta$ in Table 1)	0.016	0.0088

Detector Requirements

We make the following assumptions:

1. In the future, data collection will only be carried out with two-dimensional position-sensitive detectors.
2. The time-averaged neutron flux at the sample is taken as 3×10^8 n/cm² · sec. This figure is 100 times the flux at the Brookhaven HFBR protein crystallography beam line H3A, and is in the range of those estimated for the proposed advanced pulsed and steady-state sources (vide ante). The horizontal and vertical beam divergence is taken as 0.2° so as to match the measured mosaicity of D₂O-soaked protein crystals.
3. The position-sensitive detector is assumed to be about 200 mm x 200 mm in area and to have 256 (horizontal) x 128 (vertical) pixels. This represents no advance in resolution on existing detectors which are limited by the range of fission fragments and by possible wire spacings. Simultaneous coverage of the full 2θ range of interest can be achieved only by using an array of detectors, each element having the specifications of the present Brookhaven position-sensitive detector: Such de-multiplexing is, in any case, essential to achieve the necessary count-rate capability (see below).

For a protein crystal with a 100 Å unit cell and instrument parameters as in arrangement II in Table 2 we thus arrive at the following figures for a steady-state source:

Intensity of medium-strength reflection	$3 \text{ n} \cdot \text{sec}^{-1}$
Time for 10^3 counts ($\sigma(I)/I = 3.16\%$)	333 sec
Time for 90° crystal rotation	28 h
Total count rate (reflections plus background) for detector array	10^6 sec^{-1}
Total count rate per detector	$2.5 \times 10^5 \text{ sec}^{-1}$

The total data collection time for such a crystal would be about 36 h with a 4-sector detector and about 6 days with a single 200 mm x 200 mm detector capable of movement on the 2θ -arm of the diffractometer. In the first case two crystal orientations are necessary and sufficient to survey one asymmetric unit of the reciprocal lattice in two crystal rotations of $\sim 90^\circ$ and $\sim 30^\circ$ about the ω -axis. In the second case about 8 crystal orientations are needed.

The gain factor for a pulsed source depends on the wavelength band which can be utilized in the experiment. For our 100 Å unit cell this band extends from about 1 Å to 4 Å, and we have seen that under these conditions the gain over a benchmark steady-state experiment is between 1 and 5. This is the factor by which the time-averaged intensity of a given reflection exceeds the intensity recorded in the steady-state experiment. The gain in the time required for a complete three-dimensional data collection is much less: For a given crystal orientation only a limited part of reciprocal space is illuminated. An array of detectors must be used which covers scattering angles 2θ from 0° to 90° : $2\theta_{\max}$ is greater than for a steady-state source, because in the present case small-spacing reflections are measured with longer wavelength neutrons. At least 9 detectors of the dimensions of the present BNL position-sensitive detector are needed to cover this angular range in order to keep the angle of incidence of the detectors acceptably small. Under these conditions the illuminated volume which is shown shaded in the figure is only 9.1% of the volume of the 1.6 Å limiting sphere. The shape of the shaded volume does not lend itself to an efficient filling of reciprocal space, and many, (typically about 30), crystal orientations are needed for the collection of a complete data set.

As noted above, however, experiments in which all reflections must be measured over a range of wavelengths, for example in order to make extinction corrections, or for anomalous dispersion phasing, can be carried out very conveniently at a pulsed source by giving a complete rotation to the specimen crystal.

Apart from these differences in data-collection strategy there are important differences between the two sources as regards detector counting losses and data acquisition and storage.

1. Dead Time

(i) Steady-State Source. For a 10% counting loss a total count rate of $2.5 \times 10^5 \text{ sec}^{-1}$ per elementary detector requires a dead time of 400 nsec. This is possible with today's technology.

(ii) Pulsed Source. The mean count rate is G times that for (i) above, but this rate is far from uniform in time. It will peak fairly sharply at that time in the pulse which corresponds to the arrival of 1.5 Å neutrons. The effective dead time of a detector for an intensity-modulated signal is not very sensitive to the exact nature of the modulation; in the present case we would estimate it to be about 3 times greater than the constant-rate dead time. For 10% losses the maximum permissible dead time for each of the 9 elementary detectors is thus about

$$\frac{9}{3G} \times \frac{0.1}{10^6} \text{ sec} = \frac{300 \text{ nsec}}{G}$$

and this will be difficult to achieve.

2. Data Processing

A complete data set from a 100 Å unit cell measured at 1.6 Å resolution contains about 2×10^6 reflections. Each reflection must be sampled at about $6 \times 6 \times 10 = 360$ sampling points ($X \times Y \times \omega$ for a steady-state source, $X \times Y \times \lambda$ for a pulsed source). Some form of three-dimensional profile-fitting is desirable to extract the maximum information from the measurements, so that about 720 Mbytes of information must be stored for a complete data set.

The amount of storage space which has to be available in a macroscopic time interval differs greatly for steady-state and pulsed

sources. In a steady-state experiment the specimen crystal is rotated at about $0.05^\circ/\text{min}$, and at any instant only those reflections are active which lie within about 1° of the Ewald sphere, i.e., less than 1% of the total. With a pulsed source the shaded volume element is swept through in each pulse interval and the instantaneous storage requirements are vastly greater.

Conclusions

It is expected that data collection would proceed at about the same rate with either of the two proposed advanced sources. For a steady-state reactor existing state-of-the-art devices would be adequate and data processing and storage would present no great problems. For a pulsed source some detector development is necessary; instantaneous storage requirements would be perhaps an order of magnitude greater than for a steady-state source, but this should be economical to provide by the time the proposed source would be operating.

Neutron Guides for Pulsed Sources

In order to resolve the diffraction orders from proteins with unit cells of approximately 100 Å, flight paths of the order of $> 20 \text{ m}$ are required. This will involve transporting neutrons through neutron guide tubes. Conventional neutron guide tubes are made of nickel-coated glass plates. Nickel is selected as the reflecting material because it has a critical angle per unit wavelength of $0.097^\circ/\text{Å}$, which is the largest value of all the readily available materials. Neutrons incident on the walls of the guide tube up to an angle equal to the critical angle will be reflected, while the rest will be lost from the beam due to absorption processes. For neutrons with larger wavelength ($> 3 \text{ Å}$) the critical angle will be sufficiently large to transport almost all the usable neutrons to the spectrometer. For thermal neutrons, however, the critical angle will be very small and a substantial flux of neutrons will be lost in the neutron guide tubes. The critical angle of nickel for 1.5 Å neutrons is only 0.15° .

A supermirror is composed of a large number of multilayer monochromators deposited on the same substrate. The d-spacing of these

monochromators will be varied in small increments so that neutrons incident at different angles can be reflected. The effective critical angle of a supermirror will be determined by the smallest d-spacing in the composite. Since multilayer monochromators with d-spacing as small as 70 Å and a reflectivity of 95% have been made with an RF sputtering system, it should be possible to make a supermirror with a critical angle three to four times that of nickel. The gain factor in neutron flux transported by a supermirror guide tube over that of a nickel-coated guide tube is proportional to the ratio of solid angles of the beams transmitted by the guides which, in turn, is proportional to the squares of the ratios of critical angles. A supermirror guide tube will therefore result in an order of magnitude greater flux.

In addition, supermirrors can be bent in such a way that the sample is not directly exposed to the neutron source, therefore reducing the strong background of gamma rays.

The neutron beam will undergo a number of reflections inside the guide. For a 25 m long guide with an inner dimension of 2.5 cm, a neutron beam of 1.5 Å will be reflected a maximum of eight times from the supermirror walls. For this number of reflections, the usefulness of supermirrors will decrease for low values of reflectivity. It is therefore desirable to have a reflectivity greater than 95% throughout the angular range of the multilayer. The following factors adversely affect the reflectivity of a multilayer monochromator: (1) surface roughness of the substrate, (2) disorder in the multilayer, i.e., deviation in the thickness of a film from the desired value, and (3) diffusion of materials between the layers.

In order to produce supermirrors of the required high reflectivity, a pilot research program should be undertaken now to solve the technical problems associated with the manufacture of these devices.

Liquids and Amorphous Materials

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A. Introduction

In this section we address three areas of potential interest: Multicomponent glasses and liquids, amorphous magnetic materials, and liquid helium-3. In each case we identify experiments that call for the next generation of neutron source.

Because of space and time limitations we have not considered topics such as collective (non-magnetic) excitations in liquids and amorphous materials, isotope substitution inelastic scattering experiments on multicomponent systems, kinetic studies, spin excitations at the boundary layer between a magnetic and a non-magnetic phase, inelastic studies of paramagnetic fluids, and critical scattering: no doubt there are other topics, hopefully covered elsewhere in these proceedings, which we have not thought of at all.

Certain types of neutron scattering experiment on liquids and amorphous materials can be performed satisfactorily using existing or proposed instruments at the present generation of reactors and pulsed sources. This applies to both diffraction and inelastic scattering studies, and includes much of the work on liquid ^4He . It is arguable, however, that systematic, careful, inelastic scattering measurements, on liquids in particular, would benefit from higher performance instruments such as those proposed for the next generation sources. One such type of study would extend existing measurements well into the high- ω wings of the scattering function, where potential-dependent effects manifest themselves more clearly: the problem here is that single scattering is weak and multiple scattering must be reduced, perhaps by using significantly smaller samples. Another type of study involves measurements at a variety of points in the phase diagram, in certain cases extended to relatively large values of Q , of order $10\text{-}20 \text{ \AA}^{-1}$.

B. Multi-Component Glasses and Liquids

1. Scientific Opportunities

Neutrons possess several advantages over x-rays for the precise determination of the structure factor of simple liquids and the partial structure factors in multicomponent glasses and liquids:

- (1) A number of elements have isotopes with different neutron scattering lengths.
- (2) Most elements have relatively small absorption cross sections, enabling one to study structure in the bulk, rather than thin foils.
- (3) Neutron scattering lengths are independent of Q .
- (4) Total, elastic and inelastic scattering experiments may be performed.
- (5) Some low- Z elements have relatively large scattering lengths.
- (6) A large range of wavelengths can be extracted both from spallation sources, and from reactors with hot and cold sources.

Neutron diffraction has provided a powerful tool to elucidate the structure of liquids and glasses. Short-wavelength neutrons from spallation sources or hot sources in conventional reactors permit the extension of the diffraction data to large Q values (as high as $50 \text{ \AA}^{-1}$). This is, in principle, also possible with high-energy x rays or γ rays, but the strong decrease of the scattering amplitudes of x rays with increasing Q makes precise intensity measurements at large Q very difficult. In contrast, the Q -independence of the coherent scattering lengths of neutrons facilitates greatly the conversion of the scattering data to $S(Q)$. As a consequence, the Fourier transforms of the structure factor, deduced from neutron data, show fewer ripples at small r values than the corresponding x-ray data.

The availability of isotopes with different scattering lengths for a particular element allows us to evaluate the partial structure factors in multi-component systems. If n elements are present in the liquid or glass, we have to determine $n(n+1)/2$ partial structure factors $S_{ij}(Q)$, whose Fourier transforms, the radial distribution functions, represent the number of j -type atoms per unit volume about an i -type atom. These functions provide information about the topological and chemical short-range order.

The most suitable candidates for the isotope substitution method are elements which contain isotopes with both positive and negative scattering lengths: examples are Ni, Ti, Dy and W. In such a case, it is possible to prepare a

mixture of isotopes such that $\bar{b}_i = 0$, a so-called "null" element; one may also prepare mixtures such that $\langle \bar{b} \rangle = 0$, e.g. the so-called "zero"-alloys. In the latter case, it is possible to determine directly the chemical short-range order in liquids and glasses.

In general, the spread in scattering length for isotopes of a particular element is rather small. As a consequence, the change in the total scattering pattern through isotopic substitution will also be small and precise intensity measurements (with a statistical error $< 0.1\%$) are required in order to deduce meaningful partial structure factors. In the case of binary systems, we have to determine three partial functions $P_j(Q)$ which are related to the three total functions $J_i(Q)$ by:

$$[J(Q)] = [V] [P(Q)] .$$

If we introduce the figure of merit T defined as

$$T = ||V|| ||V^{-1}||$$

where $||V||$ is the norm of the matrix $[V]$, then it can be shown that the relative error in P is

$$\Delta P/P \leq T \Delta J/J.$$

The figure of merit T has been found to lie in the range between 30 and 300 for metallic glass systems whose partial structure factors have been evaluated so far. Assuming $T = 100$ we must measure $J(Q)$ to better than 0.1% to limit the error in P to less than 10% .

The field of liquid structure factor determination also takes full advantage of the isotopic substitution technique. The following areas are active:

- o Liquid Alloys: The goal in the determination of the partial structure factors of liquid alloys is to compare them with the corresponding functions in amorphous alloys of similar composition.
- o Molten Salts: The aim is to obtain information on the interatomic pair potentials and for this purpose high accuracy in the low- Q region is essential.
- o Aqueous Solutions: At the lowest level, first-order differences (using 2 samples) are used to obtain the hydration around the specified ion/atom. At the next level, second order differences (using 3 or more samples) are necessary for ion partial structures. Future work is likely to move towards more dilute concentrations. There is also a move to use isotopes of elements

such as C or N where the difference in scattering lengths is small. In all these applications high fluxes are needed to give the required statistical accuracy, of order 0.1% or better.

- o Hydrogenous Materials: Another form of substitution is that of hydrogen or deuterium, which can be applied both to hydrogenous molecules and to hydrogen-bonded systems. For molecular systems, a large Q range is essential: the high- Q region to measure the intermolecular terms, the low- Q region to probe the intermolecular interactions.

Because of the relatively great penetration of neutrons, it is generally possible to select suitable container materials for liquid samples. However, when high temperatures are required, the reactivity of the liquid with the container and the environment may pose great technical difficulties. In this case, a short counting time may be necessary, requiring high total counting rates. This can be accomplished by increasing the number of incident neutrons on the sample, and by counting as many of the scattered neutrons as possible by increasing the number and area of the neutron detectors. An example of high-temperature studies would be the evaluation of the partial structure factors of liquid metallic alloys which can be retained in the glassy state by rapid quenching. Many of these alloys contain highly reactive Ti or Zr, and a short exposure time, implying a very bright neutron source, would greatly facilitate such experiments.

High-pressure studies of liquids require complicated experimental set-ups. The use of 90° scattering with white beams from spallation sources permits significant reductions in the scattering due to the sample environment. For studies of amorphous samples, a scattering angle between 30° and 50° would be desirable in order to cover a large range in Q , say from 1 to $50 \text{ \AA}^{-1}$. This requires very narrow collimation of the incident and scattered beams to eliminate the scattering from the high-pressure cell.

The requirements for large neutron fluxes and high counting efficiency of the scattered neutrons become even more severe when one combines high temperatures with high pressures. Examples include a study of the pressure dependence of the structure factor of liquid metals at different temperatures. Such experiments would shed some light on the applicability of the Kirkwood superposition principle, which states that the triplet correlation function is a product of three pair correlation functions.

High fluxes would also help in the investigation of gases where the structure factor (especially in the low- Q region) leads to information on the virial coefficients, and variations with density give information on the triplet effect.

It has been pointed out before that $S(Q)$ is not directly accessible from scattering measurements but must be deduced from the scattering data through several steps of correction procedure. The one that causes most concern corrects for departures from the static approximation. It is usually called the Placzek correction and is largest for light elements. It would be of interest to measure $S(Q, \omega)$ for a glass or liquid containing a light element, say liquid Li or glassy Ni-B, over as large a range of Q and ω as possible. The data would permit us to evaluate $S(Q)$ without making any prior assumption about the correction for inelastic scattering such as is made in standard neutron diffraction measurements.

Polarization techniques offer the important possibility of efficient separation of coherent and incoherent scattering. The losses of intensity involved in polarization of incident and scattered beams require very high fluxes. Polarized beams can also be used for looking at magnetic scattering, especially in amorphous materials.

On pulsed sources this requires the development of white-beam polarizers. This technique can also be extended, in principle, from elastic (total) to inelastic scattering experiments. Such an instrument is being developed for the SNS.

2. Instrumentation

The ILL experience has demonstrated what improvements in instrument design can do to the efficiency of neutron use at reactor sources. The D4 liquids diffractometer at the ILL has been rebuilt to give a very high count rate. This has been achieved mainly by increasing the incident flux with a focussing monochromator and by employing two multidetectors to increase the detection efficiency. These improvements have reduced counting times by an order of magnitude over the previous design. The only further improvements possible for such instruments are a factor 4 with a new high-flux reactor such as ORNL's HFIR-II design and a further gain due to replacing the 2 detectors with a single 160°-wide detector, but this comes at the expense of increased background.

The main drawbacks of such an instrument are those inherent to reactor instruments. Firstly, there is the restricted Q range, for example, $Q_{\max}$ is $\sim 17 \text{ \AA}^{-1}$ at $\lambda = 0.7 \text{ \AA}$ (assuming that the instrument is on a hot source). Higher Q values can be achieved with shorter wavelengths but at the expense of reduced flux. Secondly, the corrections for inelastic scattering are difficult to calculate, especially for light elements. Instruments on pulsed sources do not suffer as much from these problems. The use of short wavelengths and low scattering angles provides the large Q range and makes the inelasticity corrections less problematic. Experimental work on silica at both reactor and pulsed sources has shown that these corrections are unreliable above about 90° . For samples containing elements of lower mass, the upper limit in angle may be as low as 20° . The solution is therefore to concentrate the detectors at small scattering angles to reduce the corrections, and to extend the Q range at a particular angle by using higher energy neutrons. As an added bonus, the corrections are more reliable at high energies.

The instrument should be able to measure in the Q range from $0.05 \text{ \AA}^{-1}$ to at least $50 \text{ \AA}^{-1}$. The upper limit ensures adequate resolution in real space when Fourier transforming the data. Angular resolution at around 30° scattering angle ($1 < Q < 80 \text{ \AA}^{-1}$) should be $\leq 1\%$. In order to take full advantage of the higher energy neutrons, the detector must also have sufficient efficiency in that energy range. This is possible by using ^6Li loaded glass scintillators - for example, a 5mm thick scintillator has an efficiency of 70% at 1eV ($0.3 \text{ \AA}$) and 10% at 100 eV ($\sim 0.03 \text{ \AA}$).

Comparisons of flux between the two types of source are complicated by the variation in count rate with scattering angle in the pulsed case. However, while a well designed reactor instrument could approach the count rate of pulsed instruments over a limited low- Q range, the advantages outlined for short wavelengths, which are unavailable at reactors, should lead to better data, and hence a more reliable interpretation of the data, if a pulsed source is used.

The installation of a correlation chopper on a total scattering spectrometer will allow the measurement of inelastic scattering. The full $S(Q, \omega)$ can be used to calculate the "true" $S(Q)$ for Q corresponding to elastic scattering at a particular scattering angle, while a direct summation of the data at that angle gives the result obtained on a total scattering instrument but reduced in flux by the duty cycle of the chopper ($\sim 1/2$). A comparison of the two intensities can then be used as a check on the accuracy of the Placzek correction procedure.

C. Amorphous Magnetic Materials

Studies of amorphous magnetic materials and the related subject of spin glasses present some of the most important and challenging problems in magnetism today. This interest is increasing rapidly and may be expected to accelerate substantially in the future. These studies involve several aspects, including

- o Determination of the degree of elasticity of the spin-glass order parameter.
- o Measurement of the dispersion of propagating spin excitations in ferromagnetic amorphous alloys.
- o Linewidth studies of spin excitations as a function of Q and T .
- o Investigation of possible Stoner band continuum effects in amorphous alloys at relatively large Q ($Q \approx 0.5 \text{ \AA}^{-1}$).
- o Investigation of co-existent (or re-entrant) ferromagnetic and spin glass order in amorphous alloys. This involves simultaneous high resolution of the dynamic (spin-wave) response and the elastic (or quasi-elastic) spin glass order parameter.
- o Measurement of spin relaxation effects in spin-glass systems in real time.

There are a number of experimental features and requirements for these studies, some of which are rather unusual. In general scattering data must be obtained in the small- Q region, which is the only region of phase coherence for amorphous alloys. High-energy resolution is required and low- Q capabilities are necessary to examine the very-long-wavelength behavior of these systems. A prototype problem might be described in the following terms:

1. Spin-wave excitations propagate in the ferromagnetic phase with dispersion $E = DQ^2$, where $60 < D < 180 \text{ meV \AA}^2$.
2. The lifetime of the spin-wave excitation needs to be determined accurately.
3. In the lower temperature region of the ferromagnetic phase, spin glass order may be present and coexist with ferromagnetism. This is manifested as a broadening of the δ -function order-parameter scattering at $Q = 0$ in the ferromagnet into diffuse scattering with Lorentzian or more complex line-shape. The degree of elasticity of this central peak is to be determined.

Typical (possibly worst-case) requirements for the scattering experiments are given below. Ideally the entire Q and E range should be accessible using a single instrument.

1. Extreme low (Q,E) region:

Minimum Q	0.02 $\text{\AA}^{-1}$
Minimum E	0.05 meV
Energy resolution ΔE	< 0.03 meV

Q-resolution, due to vertical beam divergence, of same magnitude or less than Q.

Such requirements could be met with a cold-source triple-axis spectrometer on a 10^{16} source using $E_i = 3.5$ meV, with 10' collimators throughout.

2. Medium and high (Q,E) region:

Maximum Q	0.5 - 0.8 $\text{\AA}^{-1}$
Maximum E	25 - 60 meV
ΔE	0.1 E or better, preferably ~ 0.04 E.

This implies incoming energies E_i of order 300 - 500 meV with resolution better than $0.005 E_i$. This requirement can be met with a hot-source triple axis or with the low-angle chopper spectrometer described in Section E.

As stated above, these studies optimally should be performed on a single spectrometer; however it may be necessary in practice to use separate instruments (or to exchange choppers) for the two regions of Q and E.

The source intensity is critical at all Q. At the lowest Q, resolution requirements will probably limit the available sample flux while at higher values of Q the cross section is reduced (since it varies as $\approx 1/Q^2$) and the excitations may broaden appreciably (due to conventional dynamic effects and possible Stoner-continuum effects).

In addition polarization analysis is very useful for many studies, and it should be incorporated in any instrument design. The polarization capability should ideally be available as a "pushbutton option" on a machine which is otherwise basically a non-polarized spectrometer (like IN20 at ILL for example). Significant concerns exist regarding the feasibility of polarized-beam time-of-flight spectrometers (for example at pulsed sources).

The required combination of polarization analysis, high energy and good Q resolution clearly dictate the need for a higher flux ($\approx 10^{16}$) source in order to do useful experiments, particularly on dilute magnetic systems.

D. Liquid Helium-Three

1. Elementary Excitations

A measurement of the elementary excitation spectrum of liquid ^3He is of great fundamental interest as this is the only neutral Fermi liquid available for laboratory study. The elementary excitations in liquid ^3He are of two kinds, normally number-density fluctuations and spin-density fluctuations. Neutron scattering experiments to date have shown that for $Q \lesssim 1.3 \text{ \AA}^{-1}$ the scattering function has two peaks. One peak within the particle-hole (p-h) band at low energy corresponds to the spin fluctuations, and one peak at higher energy corresponds to the density fluctuations (see Fig. 1). The peak within the p-h band is identified as the paramagnon resonance and is due to the spin-antisymmetric part of the interaction. This interaction results from a delicate balance between the interaction of pairs of particles in the triplet and singlet state respectively. The resulting interaction is very sensitive to relative changes in the two components. Theoretical analysis of the available results indicates that the system is close to a ferromagnetic instability at SVP. Due to the Fermi exclusion principle, the singlet and the triplet state should respond differently to changes in the density of the particles. Variation of the density can therefore be expected to bring about dramatic changes in the spin fluctuation spectrum and measurements at elevated pressures are thus of considerable interest.

The density fluctuations (zero-sound) mode is governed by the spin-symmetric interaction. The mode shows large positive dispersion for Q in the range $0.6 - 0.8 \text{ \AA}^{-1}$. The width of the zero-sound mode as a function of Q implies complex damping processes, probably including decay into multiple density excitations as well as multiple p-h excitations. From the variation of the Landau parameters with pressure one expects a strong pressure dependence in the zero-sound mode. The mode frequency should increase with pressure while, due to the increase in the effective mass, the p-h band narrows and shifts to lower energy. At elevated pressure the two peaks will therefore be easier to isolate in the experimental results. The coupling between the zero-sound mode and the p-h excitations should also change with corresponding variation in the width of the zero-sound mode.

An adequate facility for measurement of the shape of the paramagnon peak should meet the following specifications:

Energy resolution	5-10 μeV
Energy transfer range	0-1000 μeV
Wave-vector transfer range	0-1 $\text{\AA}^{-1}$
Flux on sample	$> 10^5 \text{ n/cm}^2/\text{sec}$
Beam size	10 cm^2

It should be noted that in extreme cases the paramagnon peak will overlap the elastic scattering from the container. In such cases the paramagnon scattering can be isolated by spin-flip analysis if a container material with no spin-flip scattering is used. This option would require a further increase in the flux by about a factor of 4. The source requirements can be estimated by comparison with the performance numbers of the chopper instrument IN5 at ILL. At 12 $\text{\AA}$ incident energy the resolution is $\sim 8 \mu\text{eV}$ and the flux on the sample is then $\sim 1.0 \times 10^4 \text{ n/cm}^2/\text{sec}$. A source of strength one order of magnitude higher than the HFR reactor at ILL is thus required but the experiment can more readily be done when an ASPUN - class source is available.

By comparison, the measurement of the zero-sound dispersion and the zero-sound broadening is less demanding and this experiment could be done either at ASPUN or at the proposed $5 \times 10^{15} \text{ n/cm}^2/\text{s}$ reactor at Oak Ridge.

2. Momentum Distributions

At wave vectors sufficiently large for the impulse approximation to be valid, measurements of the scattering function $S(Q, \omega)$ give information about the momentum distribution in the ground state $n(p)$. In ^3He at temperatures well below T_F (1.6° K), $n(p)$ is expected to have a discontinuity at the Fermi surface, $p=p_F$. This is reflected by kinks in $S(Q, \omega)$ at $\omega = \hbar Q^2/2M \pm Qp_F/M$ (see Fig.2). Observation of these by neutron scattering would provide a direct observation of the Fermi surface in liquid ^3He , analogous to measurements made in metals with electron Compton scattering.

The requirement of large Q means that measurements of this kind are especially suited to pulsed-source instruments using epithermal neutron energies. A preliminary measurement at IPNS with $E_i = 0.26 \text{ eV}$ yielded an approximate estimate of the width of $n(p)$, i.e. the mean kinetic energy per particle. However,

the very high absorption cross section of ^3He means that present pulsed sources cannot provide sufficiently intense beams for high-resolution measurements to be carried out. To make a proper observation of the kinks in $S(Q, \omega)$, we estimate that about five resolution elements will be needed on the relatively straight sections of the curve on either side of the kink. For $Q = 15 \text{ \AA}^{-1}$, the curve in Fig. 2 implies that a resolution $\Delta E = 1.5 \text{ meV}$ will be required. This resolution must be accompanied by sufficient flux on sample to provide adequate statistics for a detailed shape analysis of $S(Q, \omega)$. An appropriate machine is the Ultra-High Resolution Chopper Spectrometer¹. This instrument is designed for exactly the resolution required, $0.005 E_f$, but times must be scaled up by 2 appropriate for an E_f of 0.25 eV instead of 1.0 eV . In conjunction with the parameters of moderator A (Crawford et al.²), this gives the following parameters:

$$E_f = 0.25 \text{ eV}$$

$$\Delta E = 1.25 \text{ meV}$$

$$\bar{\phi}_s = 1.1 \times 10^5 \text{ n/cm}^2.\text{sec}$$

This flux is 20 times that available in the IPNS experiment, so that statistics should be 4-5 better, certainly adequate for shape analysis.

In addition to studies of the effect of pressure, addition of ^4He , etc. on the Fermi surface, we may anticipate the eventual capability to make measurements on polarized ^3He . (This will require high magnetic fields and very low temperatures; however, measurements on solid ^3He at 0.5 mK and 0.1 T are already being carried out at IPNS.) In partially polarized ^3He one may expect two Fermi surfaces, for the majority and minority spins, both of which should show up in the $n(p)$ measurement.

Neutron measurements at moderate to high Q have established the existence of the Bose condensate in ^4He . The measurement of the effects of temperature, pressure, and addition of ^3He on the condensate fraction is currently an active area of investigation at present sources. Looking ahead to technological developments, we can anticipate $n(p)$ measurements on spin-polarized hydrogen, another monatomic Bose system. In addition to measurements of broad features of $n(p)$ such as the kinetic energy, it may be possible to ascertain the existence of a Bose condensate in spin-polarized hydrogen.

E. Low-Angle Chopper Spectrometer

This instrument is a chopper spectrometer designed to measure collective excitations in isotropic systems at low wave vectors and moderate energy transfers. This situation applies, for example, to spin waves as well as Brillouin excitations in the "first zone" in amorphous metals, and to fractal excitations in glassy solids. In a typical amorphous metal we may have a spin wave energy $\hbar\omega = DQ^2$ where $D \sim 160 \text{ meV/\AA}^2$; if, for example, $Q = 0.5 \text{ \AA}^{-1}$, then $\hbar\omega = 40 \text{ meV}$ requiring an incident energy of 1000 meV and a scattering angle $\phi = 0.61^\circ$. A Brillouin excitation may be observed at $\hbar\omega = v_s Q$ where v_s typically $\sim 4000 \text{ m/sec}$; if, for example, $Q = 0.5 \text{ \AA}^{-1}$, then $\hbar\omega = 14 \text{ meV}$, requiring an incident energy of 250 meV and a scattering angle $\phi = 2^\circ$.

We set the requirement that the energy resolution be

$$\Delta E \sim 0.005 E_i, \quad (1)$$

so that $\Delta E = 5 \text{ meV}$ and 1.25 meV respectively in the two examples above. This requires an initial flight path $L_1 \sim 20 \text{ m}$. An optimized instrument has the final flight path $L_3 \gg L_1$ but this leads to unrealistically large detector arrays and shielding. In fact $L_3 \sim L_1$ is a reasonable compromise. In order to provide the capability to measure down to much smaller values of Q and $\hbar\omega$, we envisage the instrument placed at a liquid H_2 moderator (moderator C of Crawford *et al.*²). In this case measurements at different (Q, ω) ranges could be accommodated by changing the chopper to one with an appropriate slit width and curvature, perhaps a 6-hour change. Over a limited range this could instead be accomplished with a change in rotor speed. The instrument could be combined with the Ultra-High Resolution Chopper Spectrometer¹, which has $L_1 = 20 \text{ m}$, $L_3 = 7 \text{ m}$, by providing a longer scattered flight path with $L_3 = 20 \text{ m}$ at low angles. A similar instrument but with lower resolution has been proposed for SNQ by Reichardt.³

Neglecting the finite thicknesses of sample and detector, which have a negligible effect on resolution for 20 m flight paths, the energy resolution is given by⁴

$$\Delta E = m \left\{ \left(\frac{v_i^3}{L_1^3} + \frac{v_f^3 L_2^2}{L_1^3 L_3^2} \right) \Delta t_s^2 + \left(\frac{v_i^3}{L_1^3} + v_f^3 \frac{(L_1 + L_2)^2}{L_1^3 L_3^2} \right) \Delta t_o^2 \right\}^{\frac{1}{2}}, \quad (2)$$

where Δt_s and Δt_c are the pulse widths at source and chopper respectively. Intensity is optimized when the two terms in the above expression are equal. For $v_f \sim v_i$ this occurs when

$$\Delta t_c = \frac{L_2 + L_3}{L_1 + L_2 + L_3} \cdot \Delta t_s \quad (3)$$

and then

$$\frac{\Delta E}{E_i} = \frac{2\sqrt{2} (L_2 + L_3)}{L_1 L_3} v_i \Delta t_s \quad (4)$$

In the slowing down region, Fig. 2 of Ref. 2 shows that

$$\Delta t_s \approx 2 E_i (\text{eV})^{-\frac{1}{2}} \mu\text{sec} = 2 (13,831/v_i (\text{m/sec})) \mu\text{sec}. \quad (5)$$

Putting $L_1 = L_3 = 20 \text{ m}$, $L_2 = 1 \text{ m}$, we get

$$\begin{aligned} \frac{\Delta E}{E_i} &= 2\sqrt{2} \left(\frac{21}{20 \cdot 20} \right) 2 \cdot 10^{-6} \times 13,831 \\ &= 0.0041. \end{aligned} \quad (6)$$

At much lower energies, down to 1 meV, $v_i \Delta t_s$ increases by a factor 2, so $\Delta E/E_i$ increases to 0.008. This is still approximately consistent with the requirement of Eq. (1).

The wave-vector resolution in the scattering plane is given by⁴

$$\begin{aligned} \Delta(Q^2) &= 2 \left(\frac{m}{h} \right)^2 \left\{ \left[\frac{v_i^2}{L_1} (v_i - v_f \cos \phi) + \frac{v_f^2 L_2}{L_1 L_3} (v_i \cos \phi - v_f) \right]^2 \Delta t_s^2 \right. \\ &+ \left. \left[\frac{v_i^2}{L_1} (v_i - v_f \cos \phi) + v_f^2 \frac{(L_1 + L_2)}{L_1 L_3} (v_i \cos \phi - v_f) \right]^2 \Delta t_c^2 + [v_i v_f \sin \phi]^2 \Delta \phi^2 \right\}^{\frac{1}{2}}. \end{aligned} \quad (7)$$

For small values of ϕ and $v_f \sim v_i$ we get

$$\Delta(Q^2) = 2 \left(\frac{m}{h} \right)^2 \left\{ \left[\frac{v_i^2}{L_1} (v_i - v_f) \left(1 + \frac{L_2}{L_3} \right) \right]^2 \Delta t_s^2 \right.$$

$$+ \left[\frac{v_1^2}{L_1} (v_1 - v_f) \left(1 + \frac{L_2}{L_3} \right) \right]^2 \Delta t_c^2 + \left[v_1^2 \phi \right]^2 \Delta \phi^2 \}^{\frac{1}{2}}. \quad (8)$$

If Eq. (3) is satisfied, the first two contributions to $\Delta(Q^2)$ are equal and it is reasonable to make the third contribution equal the same value:

$$\begin{aligned} \phi \Delta \phi &= \frac{(v_1 - v_f)}{L_1} \left(1 + \frac{L_2}{L_3} \right) \Delta t_s \\ &\approx \frac{1}{4\sqrt{2}} \frac{\Delta E \cdot \hbar \omega}{E_1^2}. \end{aligned} \quad (9)$$

For the case discussed above ($E_1 = 1000$ meV, $\hbar \omega = 40$ meV, $Q = 0.5 \text{ \AA}^{-1}$), $\phi = 0.61^\circ$, and Eq. (9) gives $\Delta \phi = 0.16^\circ$, implying a detector of width 5.5 cm centered at 20 cm from the beam. To increase counting rate, circular detectors centered on the beam line are used so that the total detector solid angle is

$$\begin{aligned} \Delta \Omega &= 2\pi \phi \Delta \phi \\ &= 4\pi (\frac{1}{2} \phi \Delta \phi) \\ &= 4\pi \times 0.145 \cdot 10^{-4} \text{ ster.} \end{aligned} \quad (10)$$

Eqs. (3), (8), and (9) give

$$\begin{aligned} \Delta(Q^2) &= 2 \left(\frac{m}{\hbar} \right)^2 v_1^2 \frac{\sqrt{3}}{4\sqrt{2}} \frac{\Delta E \cdot \hbar \omega}{E_1^2} \\ &= \sqrt{\frac{3}{8}} k_1^2 \frac{\Delta E \cdot \hbar \omega}{E_1^2} \\ \frac{\Delta Q}{Q} &= \frac{1}{2} \frac{\Delta(Q^2)}{Q^2} = \sqrt{\frac{3}{32}} \left(\frac{k_1}{Q} \right)^2 \frac{\Delta E \cdot \hbar \omega}{E_1^2}. \end{aligned} \quad (11)$$

For the case under discussion, $k_1 = 22.0 \text{ \AA}^{-1}$ and

$$\frac{\Delta Q}{Q} = 0.10 \quad (12)$$

which is consistent with the effective energy resolution, $\Delta E/\hbar \omega \sim 0.1$.

The source intensity in the slowing down region is given by Fig. 1 of Ref. 2 as $I(E) = 4.5 \times 10^{11} \cdot E_i(\text{eV})^{-1} \text{ n/ster. sec. meV}$. For the C moderator this relation holds approximately down to $E \sim 3 \text{ meV}$, below which it begins to fall off according to the Maxwellian spectrum. The flux at the sample (pulsed, monochromatic neutrons) is given by

$$\begin{aligned}\phi_s &= \frac{I(E_i)}{(L_1 + L_2)^2} \cdot E_i \cdot \frac{2v_1 \Delta t_c}{L_1} \cdot T_R \\ &= \frac{4.5 \cdot 10^{11}}{2100^2} \cdot 1000 \cdot \frac{2 \times 13831 \times 2 \cdot 10^{-6} \times \frac{21}{41}}{20} \cdot 0.5 \text{ n/cm}^2 \cdot \text{sec} \end{aligned}$$

where T_R is chopper transmission factor assumed equal to 0.5.

$$\phi_s = 7.2 \cdot 10^4 \text{ n/sec}^2 \text{sec.} \quad (13)$$

For a sample with area $10 \text{ cm} \times 5 \text{ cm}$, a scattering probability of $0.1 \Delta\Omega/4\pi$, and a detector efficiency of 0.5, the count-rate in a detector with $\Delta\Omega$ given by Eq. (10) is

$$\begin{aligned}C_d &= 7.2 \cdot 10^4 \times 50 \times 0.1 \times 0.145 \cdot 10^{-14} \times 0.5 \text{ ct/sec} \\ &= 2.61 \text{ ct/sec} \\ &= 160 \text{ ct/min} \end{aligned} \quad (14)$$

This count-rate is acceptable, especially in view of the relatively low background experienced at pulsed spallation sources. We note that the relative energy resolution given by Eq. (1), flux on sample given by Eq. (13) and counts at detector given by Eq. (14) are essentially constant over the entire E_i range of 3 to 1000 meV.

Chopper Requirements. We require a chopper with transmission $T_R > 0.5$, $\Delta t_i \approx 1/2 \Delta t_s \approx 1 \mu\text{sec} \cdot (13,831/v_1)$, chopping with high on-off ratio at energies up to 1000 meV. This requires a slit width d , radius R and frequency ν given by

$$\Delta t_c = \frac{d}{2R \cdot 2\pi\nu} = 10^{-6} \cdot (13,831/v_1) \text{ sec.} \quad (15)$$

The increase in pulse width due to the "lighthouse" effect is

$$\Delta t_l = \frac{a}{2\pi\nu} \quad (16)$$

where α is the incident collimation $\approx 10 \text{ cm}/21 \text{ cm} = 4.8 \times 10^{-3}$. Presently the maximum feasible chopper speed is $\nu \sim 600 \text{ Hz}$, so Eq. (16) gives

$$\Delta t_g = 1.27 \cdot 10^{-6} \text{ sec}$$

which will degrade the resolution by a factor $(1.27^2 + 1)^{\frac{1}{2}} = 1.62$ at $v_1 = 13,831 \text{ m/sec}$, decreasing for lower energies. Unless higher performance choppers can be developed, it will be necessary at the highest energies to decrease the incident collimation with a Soller collimator, causing some loss in beam intensity. For $\nu = 600 \text{ Hz}$, the ratio $(d/2R) = 3.8 \times 10^{-3}$, which can be easily accomplished with, for example, $2R = 20 \text{ cm}$, $d = 0.076 \text{ cm}$. Separator plates of boron-aluminum composite now used at IPNS have thickness $t = 0.051 \text{ cm}$, so $d/(d + t) = 0.6$ consistent with the requirement $T_R > 0.5$.

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Fig. 1 Scattering function from liquid ^3He at $Q = 0.9 \text{ \AA}^{-1}$ [K. Sköld and C. A. Pelizzari, *Phil. Trans. Roy. Soc., Ser. 13*, 290, 305 (1980)].

Fig. 2 Calculated scattering function $S(Q, E)$ from liquid ^3He at $Q = 13.85 \text{ \AA}^{-1}$, with (●●●●) and without (-----) the effects of experimental resolution currently available at the LRMECS spectrometer at IPNS. The insert on the left shows a theoretical momentum distribution $n(k)$ from which the scattering function was derived: the discontinuity in $n(k)$ at $k = 2k_F$ produces a kink in $S(Q, E)$ at $E = \hbar^2 Q k_F / M$. [P. E. Sokol, K. Sköld, D. L. Price and R. Kleb, *Phys. Rev. Letters* 54, 909 (1985)].

POLARIZED NEUTRONS AND THEIR APPLICATIONS

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I. INTRODUCTION

The neutron spin can be exploited in a variety of ways in neutron scattering to provide additional information about the scattering system or to allow the construction of devices useful in scattering experiments. The full exploitation of the neutron spin has not been achieved because devices to produce polarized neutrons and to analyze the polarization of the scattered beam have resulted in large intensity losses. Fortunately, there have been great advances in the technology of polarizing devices in recent years so that the use of polarized neutron beams is now increasing rapidly. Even with these advances, most polarized neutron experiments are severely limited by the final counting rate. The construction of new sources with fluxes 5-10 times higher will result in a significant increase in the use of polarized neutrons with a corresponding gain in scientific information.

The full characterization of a neutron involves the specification of its momentum vector and its spin vector. On scattering, not only the direction and magnitude of its momentum may change, but the direction of the spin vector may change. The most complete information about the scattering system is obtained by fully characterizing the scattered neutron including the direction of the final spin vector. This vector polarization analysis has, until now, been mostly utilized in the analysis of transmitted beams in studies of magnetic domains. However, work is now in progress at the ILL which will result in a vector polarization analysis diffractometer for Bragg scattering.

Much information about the scattering system is retained by performing a simpler experiment in which the component of the final polarization in the direction of the initial polarization is observed. This type of experiment commonly is known as polarization analysis. The beam polarization is described by the scalar quantity

$$P = p^+ - p^- ,$$

where $p^\pm$ is the probability that the $(\pm)$ neutron spin state is occupied. These states are defined relative to the direction of the magnetic field at the sample position (z direction). The scattering is then fully defined by four cross sections, σ^{++} , σ^{+-} , σ^{-+} , and σ^{--} . By introducing polarization sensitive devices and spin flippers before and after the sample, all four of these cross sections can be measured. This turns out to be an enormously useful thing to do because one can achieve complete separation of magnetic scattering from almost all types of nuclear scattering, or in other systems can separate the nuclear spin incoherent scattering from coherent nuclear scattering. It is this type of experiment applied to inelastic scattering which will be much more scientifically productive with the advent of neutron sources with higher flux. Table 1 illustrates how the various cross sections can be separated. Here σ_{NSI} is the nuclear spin incoherent cross section, σ_{NC} is the nuclear coherent cross section, σ_{NCM}^z is a cross term involving the product of nuclear coherent amplitudes and the z component of magnetic amplitudes, and σ_J is a "junk" cross section which includes instrumental background and spurious scattering from the sample. In general, the cleanest separation of the transverse magnetic scattering is obtained by measuring the spin-flip scattering in the two field directions and subtracting one from the other. If the "junk" is independent of the field direction, this will result in an unambiguous answer. It also doubles the experimental time while increasing the fractional statistical errors. If the "junk" cross section is well-behaved, it is frequently sufficient to measure the spin-flip scattering in only one field orientation.

Table 1. Tabulation of cross sections in a typical polarization analysis experiment. $\hat{z}$ is always parallel to H .

	Spin Flip σ^{-+}	Non Spin Flip σ^{++}
$\vec{H} \parallel \vec{Q}$	$\sigma_{\text{Mag}}^{\text{xx}} + \sigma_{\text{Mag}}^{\text{yy}} + \frac{2}{3} \sigma_{\text{NSI}} + \sigma_J$	$+\frac{1}{3} \sigma_{\text{NSI}} + \sigma_{\text{NC}} + \sigma_J$
$\vec{H} \perp \vec{Q}$	$\sigma_{\text{Mag}}^{\text{xx}} + \frac{2}{3} \sigma_{\text{NSI}} + \sigma_J$	$\sigma_{\text{Mag}}^{\text{zz}} + \frac{1}{3} \sigma_{\text{NSI}} + \sigma_{\text{NC}} + \sigma_{\text{NCM}}^z + \sigma_J$

A simpler experiment, and one less costly in neutron intensity, is frequently useful in which one polarization device is omitted either before or after the sample. For example, the spin-dependent cross sections $\sigma^+ = \sigma^{++} + \sigma^{+-}$ and $\sigma^- = \sigma^{-+} + \sigma^{--}$ can be measured in such an arrangement by removing the polarization device after the sample. Such an arrangement is useful for ferromagnetic samples and in studies of nuclear polarization.

Finally, in unpolarized neutron experiments the observed cross section is given by the sum of all four partial cross sections and all information conveyed by the neutron spin is lost.

The other type of application of polarized neutrons is the utilization of the neutron spin as a tool for the determination of the energy (or momentum) transfer in the scattering process, i.e. some kind of neutron spin modulation is used to monochromatize the incoming beam or to analyze the momentum (energy) state of the scattered beam, or to use more subtle schemes like neutron spin echo (NSE). These kinds of applications are equally useful in both magnetic and nonmagnetic scattering studies where high resolution is required.

The highest resolution can be obtained by making use of Larmor precession of the neutron spins in a homogeneous magnetic field. In reasonable fields and over reasonable distances (~ 1 kOe and 1 m, respectively) as much as 10^5 radians in the Larmor precession angle is attained. In diffraction studies this will allow the investigation of very slight distortions of lattice spacings. In the now most widely used spin modulation method, neutron spin echo, such large Larmor precession angles allow the direct observation of very small neutron energy changes in inelastic scattering. In doing this the beam does not have to be very monochromatic, and therefore NSE made high resolution experiments feasible which would be otherwise impossible because of the inverse relation between resolution and neutron intensity in conventional methods. The list of successful applications of the method includes such general areas as the study of: (a) diffusive motion of polymer molecules in solutions, (b) flexibility of macromolecules, (c) diffusion in solids (e.g., self diffusion of Ti, of adsorbates in porous structures), (d) phonon-phonon and electron-phonon effects on the lifetime of phonon excitations, (e) critical dynamics in structural and magnetic phase transitions, (f) spin

relaxation and freezing in spin glasses, (g) spin diffusion in various magnetic systems (e.g., diffusive solitons in TMMC). Developments in the near future are expected to push the resolution limit to below 1 neV, which would allow the study of dynamics of macromolecular systems on the few hundreds Å length scale, a major step towards closing the gap between neutron and light scattering experiments. Another area where strong progress is expected is construction of instruments optimized for the investigation of lifetimes of elementary excitations. The already available results on phonons and rotons in superfluid ^4He show that 1 μeV resolution is achievable in this kind of work and that there are interesting phenomena to study (e.g., the observation of the crossover from three-phonon to four-phonon decay in superfluid ^4He).

The other kind of modulation schemes (magnetic monochromators [Drabkin], fast spin flip choppers, velocity spectrum modulators [Ito]) are destined to be used in the 10^{-2} -- 10^{-3} range of $\Delta E/E$ and eventually to be combined with more classical monochromators and analyzers. This range of resolution bridges the performance of NSE and conventional methods, and they are often complementary in other respects too. For example, the Drabkin monochromator allows work with high resolution in the energy domain (as opposed to NSE working in the time domain) and therefore it is well adapted for the improved resolution study of phonon dispersion relations, hybridization between branches, Kohn anomalies, and Fermi transitions.

II. NEED FOR POLARIZED NEUTRONS

A. Magnetic Materials

The following are several important examples of magnetic scattering studies in which polarized neutrons, in particular polarization analysis, play the essential role.

1. Magnetic structure, spin density and nuclear magnetism. Whenever the magnetic scattering takes place at the same momentum transfer as the nonmagnetic scattering, polarized neutrons can be utilized to separate these cross sections unambiguously. Examples include form factor measurements of

the heavy fermion superconductor UBe_{13} and the magnetic structure determination of antiferromagnets accompanied by lattice distortions such as V_2O_3 . The recent addition of dilution refrigerators to polarized beam spectrometers has opened the new and exciting field of enhanced nuclear magnetism in compounds such as PrCu_2 . The routine attainment of very low temperatures will also allow polarization labeling of isotopes with nuclear spin. While most magnetic structures can be solved without the use of polarization analysis, this technique can be very important for complex structures such as Nd and TmS . A very pretty illustration of the value of polarization analysis is shown in Fig. 1, which shows the results of an experiment undertaken to confirm the field-induced fan phase in MnP . A glance at the data shows that the first order satellites at $(2\pm\delta, 0, 0)$ result from spin-flip scattering and are therefore associated with a modulation of the c component of the moment, while the second-order satellites at $(2\pm 2\delta, 0, 0)$ result from non spin-flip scattering and are therefore associated with a modulation of the b component of the moment. This behavior is characteristic of a fan structure.

2. Spin dynamics. There are now a large number of systems where the study of dynamical spin correlations provides the basic characterization of fundamental interactions in these materials. These include itinerant ferromagnets, mixed valence compounds, Kondo systems and spin fluctuation compounds. The study of magnetic excitations in these materials, with polarization analysis, has just begun and will be one of the active areas of polarized beam studies.

3. Low dimensional magnets. The magnetic scattering from these unique magnetic crystals, such as TMMC or CsNiF_3 , has been extensively investigated in the past several years. Many important questions, however, are yet to be answered; these include soliton dynamics in one dimensional systems and vortex contribution in two dimensional ferromagnets.

4. Disordered magnetic systems. This area of condensed matter physics may be the fastest growing topic for neutron scattering. Amorphous ferromagnets, spin glasses, mixed magnets near the percolation threshold, intercalates in graphites, fractal and random field effects are notable examples of intensive current investigations. In many cases, only the polarization analysis technique can provide unique separation of the magnetic cross sections.

5. Surface and interface magnetism. Recent experiments using mirror reflection techniques have demonstrated the potential utility of neutrons for studying magnetic interfaces, including flux penetration into superconductors. Magnetic interfaces in synthetic multilayer structures have recently been studied with low angle polarized beams. These successes imply that the general availability of long-wavelength (10-20 Å) intense beams will open many exciting areas of surface-related physics to neutron investigation. Studies of critical phenomena in surface magnetism and surface fluids also appear to be feasible. In all of these areas, more intense beams are crucial, especially in view of the fact that small samples and very narrow (and highly collimated) beams often must be used.

6. Domain topography and depolarization studies. Somewhat outside of the scattering experiments is the use of polarized neutrons for the studies of real space domain topography, the determination of spatial distribution of moment, and dynamical studies utilizing the depolarization of neutrons in transmission. An example of the last category is the recent study of $\text{Fe}_{0.7}\text{Al}_{0.3}$ spin glasses.

B. Nonmagnetic Materials

Polarization analysis can also be used for the separation of coherent and nuclear spin incoherent scattering. The latter is particularly strong for protons, but it is also sizeable for a number of other nuclei: D, Na, V, Co, Sc, etc. This separation is based on the fact that the nuclear spin scattering produces a scattered beam with $-\vec{P}/3$ polarization, where $\vec{P}$ is the polarization of the incoming beam (while the scattered beam polarization is $\vec{P}$ for coherent nuclear scattering). There is a variety of experimental situations in which one has to identify the nuclear spin incoherent (NSI) scattering contributions. The following are just a few examples.

In diffusion processes in liquids and solids the self diffusion will show up in the NSI scattering, while the coherent scattering contains both self and collective contributions. A recent successful study in this direction was the investigation of liquid Na.

The NSI scattering is a disturbing background in the determination of correlations in macromolecular systems with high proton contents (polymers, biological matter), which has to be known with a very high precision. In many cases this can only be achieved by the help of polarization analysis.

The technique can also be used in surface studies to separate adsorbate scattering from substrate scattering provided that the adsorbate contains hydrogen.

C. Nuclear Physics

The continuing effort to measure the electric dipole moment of the neutron now stands with an upper limit of about 10^{-24} cm. A more abundant supply of cold or ultra-cold polarized neutrons is needed to further reduce this limit, thereby testing several theoretical predictions.

Details of the weak interaction can be studied with two types of parity violation experiments involving the use of polarized neutrons. One involves the measurement of the asymmetry of capture γ rays resulting from capture of polarized neutrons. The other involves the observation of neutron spin rotation in transmission experiments.

III. POLARIZATION OF NEUTRON BEAMS

A. At Steady-State Sources

Considerable progress in the development of neutron polarizers has been made in recent years. For example, polarizing Heusler alloy crystals with mosaics of $\approx 0.5^\circ$ FWHM (Full Width at Half Maximum) and peak reflectivities of $\approx 50\%$ (for one spin state as compared to 80% for pyrolytic graphite) at about 15 meV have now been obtained. In addition, a new type of neutron polarizer consisting of multiple-thin film bilayers (where one of the two thin film materials is ferromagnetic) has all but replaced conventional ferromagnetic mirrors. In fact, two general types of multi-bilayer polarizing devices currently exist. One device, which has become known simply as a "multilayer", consists of a number of bilayers of a single thickness or even a distribution of thicknesses which results in a diffraction peak at a position in reciprocal space beyond that corresponding to the critical angle for total mirror reflection. The other type of multiple-thin film device has

become known as a supermirror because it is composed of a particular sequence of bilayer thicknesses which in effect continuously extend the region of total mirror reflection beyond the ordinary critical angle.

A Heusler alloy crystal can have a neutron polarizing efficiency greater than 0.99 and is useful as a monochromator over approximately the same neutron energy range as that of pyrolytic graphite. However, one potential problem in using Heusler crystals as both monochromator and analyzer on a triple-axis spectrometer is the effect of simultaneous or multiple scattering on the overall polarizing efficiency of the instrument. Another related problem is the presence of higher-order wavelength contamination.

Polarizing multilayers of Fe and Ge thin films have been made with peak reflectivities approaching 90%, polarizing efficiencies greater than 98% and bilayer thicknesses as low as 50 Å. Nevertheless, a multilayer nominally having a single bilayer thickness or "d-spacing" reflects a given energy only over a relatively narrow angular range (~ 5 min at 30 meV and ~ 25 min at 1 meV is typical). The angular acceptance of a multilayer for a particular neutron energy can in principle be increased by creating a distribution of bilayer thicknesses. However, technical problems related to the control of random fluctuations in bilayer thickness during the deposition process have not yet been completely solved, particularly in the case of multilayers with relatively small average bilayer thicknesses where the number of bilayers required to obtain a high peak reflectivity over an extended angular range increases significantly. A not unimportant advantage of using a multilayer as a polarizing element is that higher-order wavelengths are suppressed. Although Fe and Ge have scattering densities that result in polarizing efficiencies greater than 0.99 in the absence of extinction, a better matching of materials (e.g., Fe and W) is required if very high polarizing efficiencies are to be realized for a sufficiently large number of bilayers that extinction is no longer negligible.

Polarizing supermirrors have been made which extend the angular range over which nearly total reflection occurs to about twice that for an unmagnetized ordinary Ni mirror (where the critical angle $\sim 0.1^\circ$ per Å wavelength). A particularly successful polarizing "bender" consisting of a

curved Soller array of a number of Co-Ti supermirrors deposited on a Gd-Ti alloy nonreflecting and absorbing layer over glass substrates is becoming available. Further work is warranted to overcome the technical difficulties encountered in trying to extend the effective critical angle beyond twice that of a Ni mirror, especially for applications involving higher energy neutrons. The performance of multilayers, supermirrors, and Heusler crystals is summarized in Fig. 2.

Both multilayer and supermirror polarizers can be saturated in relatively low magnetic fields, of the order of a few hundred Gauss (or even much less in some cases). Another property common to both supermirrors and multilayers (at least to some degree) is that a relatively broad band of energies is reflected. A simple yet effective way to improve the relatively poor energy resolution of a multilayer or supermirror (if required) is to use it in conjunction with a crystal monochromator such as pyrolytic graphite. The pyrolytic graphite does the monochromating and the multilayer or supermirror the polarizing. However, once a neutron beam has been polarized, other more innovative and advantageous means can be used to sharpen the energy resolution. One such scheme utilizes a wavelength dependent flipping device of the kind first proposed by Drabkin. Consider a pair of polarizing multilayers or supermirrors between which a corrugated, current-carrying Al foil is inserted as shown in Fig. 3. The foil produces a small, spatially oscillating magnetic field perpendicular to the beam direction. A larger, uniform magnetic field H_0 is superimposed perpendicular to both the oscillatory field and the beam direction. Although all neutrons, independent of their velocity, precess with the same Larmor frequency in the field H_0 , only those neutrons with velocities in the neighborhood of $v_0 = a\gamma H_0/\pi$ (where γ is the gyromagnetic ratio and a is the foil spacing) satisfy the resonance condition and undergo a spin flip. Neutrons with other velocities are not flipped and are subsequently not reflected by the second multilayer or supermirror in which the magnetization direction is opposite to that of the first (this can be accomplished using a current sheet to invert the guide field direction). Together, the pair of multilayers or supermirrors and flipper produce a polarized monochromatic beam with an energy spread which can be varied simply

by changing (electronically) the length of the current-carrying section of the corrugated foil since the energy resolution $\Delta E/E$ is proportional to the reciprocal of the number of reversals in the oscillating magnetic field direction. It has been calculated that for 400 reversals, a $\Delta E/E$ of about 1% can be obtained independent of E . Because the angular divergence of the neutron beam, which is a few degrees at most, has a negligible effect on the energy resolution of the flipper (for resolution of the order cited above), this device has the remarkable property that the energy width is essentially decoupled from the angular divergence of the beam. The energy resolution and divergence of the beam can then be selected independently to suit a particular experiment.

Finally, there is a real need to develop polarizers for use at higher neutron energies, beyond the useful range of Heusler crystals. One such candidate is a ^{57}Fe crystal. Another possibility is to use a spin-dependent nuclear absorption filter as discussed in the following section.

B. Polarization of Neutron Beams at Pulsed Sources

Work on pulsed sources generally requires the handling of beams containing a broad band of energies. For this purpose, two classes of machines have been devised: those that use the cold end of the neutron spectrum ($\lambda > 3 \text{ \AA}$) and those for which epithermal neutrons are required. For the machines of the first class polarization and analysis are achieved by magnetic mirrors and supermirrors, and for higher neutron energies spin filters are being investigated. These filters are based on the spin-selective absorption or scattering of polarized neutrons. Absorption filters are effective only in a certain energy range close to an absorption line. Scattering filters based on the polarization of hydrogen polarize neutrons at all energies; however, they are most effective for very low neutron energy. This comes about because the total scattering cross section is truly spin sensitive. To achieve performance comparable to that of other polarizers, the nuclear polarization should be rather close to 100%. Table 2 indicates the present status of development. The calculated performance of the ^{149}Sm filter developed at the Rutherford Laboratory is shown in Fig. 4 (taken from reference 2). Similar information is contained in Fig. 5 for the dynamically polarized

Table 2. Status of white beam polarizer development.

Filter	Place	Magn. Field (Tesla)	Temp.	Nuc. Pol.	Comments
Absorption:					
^{149}Sm (0.09 eV) in CSMN	Rutherford	0.1	16 mK	85%	96% neutron polarization with 0.2 transmission but neutron heating
^{149}Sm in SmCo_5 ferromagnet	Rutherford	340 (int.field)	10 mK	100%	Imperfect electronic magnetization neutrons depolarized
EuS (~1 eV) ferromagnet	Brookhaven	~300 (int.field)	20 mK	60%	Neutron heating
Scattering:					
H in LMN (R.F. pumping)	ILL	1.8	0.6 K	90%	Difficult to keep stable
H in LMN (R.F. pumping)	Dubna	1.7	0.95 K	65%	V. I. Lushchikov, Yu. V. Taran, and F. L. Shapiro, Soviet Journal Nucl. Phys. <u>10</u> , 669 (1970)
H in ethyl glycol (R.F. pumping)	KENS	2.5	0.3 K	70%	Indirect ^4He cooling in development
H in YES (spin refrigerator)	Argonne	1.4	1.2 K	41%	Higher field required

proton filter on the PEN spectrometer at KENS. As illustrated in Fig. 4 and 5, a high transmittance is accompanied by a low polarization. This is a general property of spin filter devices.

IV. POLARIZED BEAM INSTRUMENTS

A. At Steady-State Sources

The classic polarization analysis (PA) instrument is a three-axis machine equipped for polarized neutron operation. However, three-axis machines are not the only ones to benefit from PA. Single crystal diffractometers have been so equipped for many years and have, with good scientific reason, functioned as dedicated PA instruments measuring electron-spin density distributions. Another current use of PA is in diffuse scattering where paramagnetic and nuclear diffuse scattering are studied. In addition, it seems reasonable to equip SANS instruments at least with polarized incident beams so that effects which generate polarization in the scattering process may be studied. Due to recent progress with polarizing mirror devices, PA has become available for spectrometers with detector banks or area detectors for broad wavelength bands above 3 Å. This opens up the possibility of using PA on time-of-flight instruments with detector sets covering a substantial solid angle. Further developments are expected to reduce the presently rather high costs of such systems and to allow their application for shorter wavelengths too.

Most neutron scattering spectrometers are optimized to be able to examine a region of (Q, ω) space with a particular resolution. Each is thus specific to a certain class of problem. In many cases, however, the feature of $S(\vec{Q}, \omega)$ which is examined either depends directly on the neutron spin state before and after scattering or must be discriminated from a background signal which has such a dependence. In such cases PA may play a useful role in identifying and characterizing the signal of interest. However, the use of PA inevitably involves an intensity penalty, and a combination of PA and conventional spectroscopy is in most cases more useful than a dedicated PA

instrument. For this combination to work efficiently, the exchange between conventional and PA modes of operation on a particular spectrometer must be easy and rapid. Thus PA should in many cases be a "bolt-on" option available on a number of standard instruments. Classic examples where this has already been implemented are cold- and thermal-neutron three-axis machines where PA often serves to identify the nature of dynamic electron-spin fluctuations and to separate coherent nuclear motion from other contributions to the scattering. With a little effort this facility could be extended for use on hot-neutron three-axis machines. For pulsed sources, the low temperature required by spin filters will make it difficult to have a quick and easy transition from unpolarized to polarized operation.

Once PA has been installed on a conventional spectrometer, it is not difficult to extend this equipment to allow for various beam modulation systems such as spin-echo or spectral modulation. Indeed, on three-axis machines, NSE systems capable of suppressing thermal diffuse scattering in measurements of Bragg peak intensities and of measuring lifetimes of elementary excitations have been tested. It is a reasonable assumption that such equipment will become standard during the next decade. This will allow a considerable extension of the range of applicability of this neutron scattering technique.

NSE spectrometers can operate both on continuous and pulsed neutron sources in basically the same manner. The major difference is that rough monochromatization of the incoming beam is achieved with a velocity selector, multilayer monochromator or, if better momentum resolution is required, by a crystal monochromator on a continuous source; while the time-of-flight method lends itself quite naturally for this purpose on a pulsed source. For most experiments neutrons within a $\sim 4 - 6$ Å wavelength band can be used on a pulsed source, while typically a 20% (~ 1 Å) band can be used on a continuous source. Therefore, roughly equal flux conditions hold for a pulsed source and a continuous source if the time average flux of the continuous source is about five times higher than that of the pulsed. However, in the study of elementary excitation line widths only a given wavelength can be used on both sources, because the NSE focusing condition can only be

fulfilled for a given scattering triangle. Therefore, in this case the average flux of the two sources has to be compared.

B. Polarized Neutron Instruments at Pulsed Sources

At the present time there are two polarized beam instruments at KENS, one at IPNS, and one to be in operation at SNS. All have different features and none has the full capability of polarization analysis and energy analysis.

The TOP spectrometer at KENS uses Co-Fe curved Soller mirrors as polarizer and analyzer (optional). It is located on a cold guide and uses a wavelength range from 3 to 10 Å, giving a flux at the sample position of $3 \times 10^4 \text{ n cm}^{-2} \text{ sec}^{-1}$. It has been successfully used in a wide variety of problems as a polarized beam diffractometer and in the PA mode for depolarization studies and diffuse scattering studies. In the depolarization studies it has been demonstrated that magnetic fluctuations in the 10^6 Hz region can be measured. The planned addition of a pulsed magnet will allow new experiments in real-time diffraction.

The PEN spectrometer at KENS was designed to produce polarized epithermal neutrons. It uses a dynamically polarized proton spin filter to produce a polarized incident beam and has detectors at short and long distances to measure both total and inelastic magnetic scattering.

The IPNS instrument uses a Co-Fe mirror as polarizer and a Be filter to produce a long-wavelength wide-band beam. It has a very narrow, highly collimated beam for magnetic depth profile studies.

The SNS instrument will have ^{149}Sm filters in dilution refrigerators for both polarization and analysis. A dual purpose chopper will serve to reduce the γ heating at the dilution refrigerator and to select a neutron energy band. Energy analysis using the cross-correlation method may be a future addition.

V. SUMMARY

High efficiency polarizing devices are now available for neutron wavelengths of 2 Å and longer. These devices can be used with equal benefit

for both steady-state and pulsed experiments provided that the short wavelength cut-off in the latter case is around 2 Å. Further development of polarizers with higher reflectivities or transmissions is needed for steady-state work at shorter wavelengths. For pulsed sources, development of broad-band high-energy polarizers with higher transmission and higher-polarizing efficiency is needed. Of the two development requirements, the pulsed source task is more difficult. The low temperatures required for the spin filters under development for spallation sources will also result in high operating cost and will present problems in reliability in long-term operation. The polarization analysis technique is of sufficient value that consideration should be given to provide this capability on all new instruments with provision to easily convert from unpolarized to polarized operation.

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Fig. 1. Polarization analysis pattern of MnP in the fan phase. First-order satellites are spin flip and second-order satellites are non spin flip.

Fig. 2. Summary of current performance of neutron polarizers.

Fig. 3. The Drabkin monochromator.

Fig. 4. Calculated performance of a ^{149}Sm (in cerous magnesium nitrate) spin filter.

Fig. 5. Calculated performance of a polarized proton filter (ethylene glycol with Cr^{V} complex). P_p is the proton polarization - typically $P_p \approx 0.65$ in the KENS operation.

High Resolution Spectroscopy

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1. Summary

In the past decade the availability of high fluxes of cold neutrons ($\lambda \gtrsim 4\text{\AA}$) has led to a significant breakthrough in the absolute energy resolution, ΔE , obtainable in neutron inelastic and quasielastic spectroscopy. Ground state rotational splittings, for example, are typically studied in bulk materials or on surfaces with resolutions of order $0.1\text{ }\mu\text{ev}$, and in favorable cases coherent relaxation processes in macromolecular systems, in particular, are observed at the level of a few neV ($1\text{ neV} = 240\text{ kHz}$). There are several reasons why cold neutrons are particularly suitable. One obvious factor is that at a given relative energy resolution $\Delta E/E_0$ (E_0 is the incident neutron energy), ΔE diminishes as E_0 becomes smaller. Related to this is the fact that low energy processes are often associated with long spatial correlations, and the necessary small momentum transfer studies are more easily performed at longer wavelength. It is becoming evident, however, that for certain problems the ability to study line shape at high resolution within a given energy window is also desirable over a wider range of momentum transfer. Here, "cool" neutron spectrometers ($\lambda \gtrsim 2.5\text{\AA}$) with resolutions of order $10\text{ }\mu\text{ev}$ are useful, for example, in the study of jump reorientational processes or in tunnelling spectroscopy.

In this context, the subcommittee defined high-resolution spectroscopy for the purpose of discussion as any technique routinely offering an absolute energy resolution $\Delta E \lesssim 50\text{ }\mu\text{ev}$. Three objectives were established:

- (i) to quantify the impact on high resolution spectroscopy of (a) a steady-state reactor having 10^{16} n.cm⁻².s⁻¹ thermal flux, and (b) a pulsed source having 10^{17} n.cm⁻².s⁻¹ peak thermal flux, 10^{15} n.cm⁻².s⁻¹ time-averaged flux, and a time-structure in the cold neutron range similar to either ASPUN or SNQ. Current instruments at the ILL, Grenoble (10^{15} n.cm⁻².s⁻¹ steady state) were used for reference;
- (ii) to identify what other gains (beyond those due to a new source) could be obtained by improving existing spectrometers or implementing new techniques; and
- (iii) to evaluate the scientific areas in which the total gains determined in (i) and (ii) above would lead to foreseeable benefits.

Three types of spectrometer currently form the basis for high resolution spectroscopy:

- time-of-flight, mostly used to study incoherent scattering with 10-50 μ ev resolution;
- backscattering, primarily used to study incoherent scattering with 0.1-10 μ ev resolution; and
- spin-echo, mostly used to study coherent scattering with 0.01-0.1 μ ev resolution. (Backscattering and spin-echo tend to be complementary.)

In addition, ultra-cold neutrons (UCN; $\lambda \geq 400\text{\AA}$) may be used for low-Q studies at high resolution (e.g., in a gravity spectrometer), and neutron spin-echo may be combined with triple-axis spectroscopy to obtain high-resolution for line-shape studies within a given $\vec{Q}$ - ω window. The last two techniques use basically only the time-averaged flux, although there is a further gain foreseeable for ultra-cold neutrons from a source with time-structure, if high reflectivity, long-wavelength monochromators can be developed.

Time-of-flight spectrometers, for which a reactor beam must be chopped, clearly gain from a pulsed source with appropriate time structure; the gain is

$$g_s = (\hat{\phi}/\bar{\phi}) (R/R_{\text{opt}})$$

where $\hat{\phi}$ and $\bar{\phi}$ are peak and average fluxes, respectively, and $R/R_{\text{opt}} \leq 1$ is the source repetition rate relative to the optimal desired repetition rate for the experiment. For ASPUN or SNQ, we expect $g_s \sim 50$ relative to current instruments, while a new reactor would gain just the factor of 10 in time-averaged flux. Further, new techniques would allow a state-of-the-art instrument to show an additional gain factor of order 3 on either source.

The highest resolution techniques, backscattering and neutron spin-echo, would each gain about a factor of 10 from a new source, either by taking appropriate advantage of time-structure on SNQ or ASPUN, or from the direct flux increase on a new reactor. In addition, new ideas for spectrometer design should lead to machine gains of order 5 on either source. The total gains to be expected (source gain x machine gain) are summarized in Table I; these figures will vary for particular experiments, but are probably valid within a factor of, say, 2 either way for a global comparison.

Technique	$\Delta E/\mu\text{ev}$	REACTOR	PULSED SOURCE
		$10^{16} \text{ n.cm}^{-2}.\text{s}^{-1}$	$10^{17} \text{ n.cm}^{-2}.\text{s}^{-1}$ (peak)
Time-of-Flight	10-50	x30	x150
Backscattering	0.1-10	x10-50	x10-50
Neutron Spin Echo	0.01-0.1	x10-50	x10-50
UCN	-0.1	x10	x1*
NSE + 3-axis	5-50	x10	x1

Table I. Expected total gains (machine x source) relative to existing spectrometers on a $10^{15} \text{ n.cm}^{-2}.\text{s}^{-1}$ steady state reactor.

*At least; may be increased.

With these general figures in mind, we next examine the scientific case. In the third section, we shall make more detailed comments on individual spectrometers and discuss which features lead to source-dependent gains, as well as the general instrumental design improvements which are possible.

2. Scientific Case

In considering how the extremely powerful new instruments discussed above may best be used, we first note certain boundary conditions. For time-of-flight or backscattering spectroscopy, current energy resolutions at a given Q are to some extent already determined by technical limitations (neutron pulse width or crystal perfection), and certain new machines will benefit primarily from greatly increased flux. (Note, however, that cool neutron spectroscopy at higher momentum transfers, in particular, will also gain significantly in energy resolution.) This flux gain may be used to increase Q -resolution when required, or it may be used to perform experiments which are currently flux (rather than resolution) limited, for example rapid measurements in real-time or on small samples. Alternatively, when only data at low Q are of primary interest (e.g. in the case of macromolecular or colloidal studies), longer wavelength may be used to improve the resolution; $\Delta E \propto \lambda^{-2}$ for time-of-flight and $\propto \lambda^{-3}$ for neutron spin-echo.

2.1 POLYMERS AND POLYELECTROLYTES

Studies of localized segmental dynamics in neutral and charged polymers provide good examples of the complementarity of these different types of gain. Inelastic neutron scattering with sub- μ ev resolution has just begun to examine the dynamics of polymer segments at previously unexplored length scales, using in particular the enormously powerful technique of contrast variation (usually by H-D substitution) to follow correlated motions of labelled sub-regions. It is possible, for example, to show that the local chain dynamics do not depend on the position in the chain by labelling different segments. Such studies, however, are limited by the fact that the scattered intensity depends on the square of the scattering volume, and higher flux is needed as finer detail is examined by labelling smaller regions. The need for much higher flux is also apparent in the new area of

polyelectrolyte studies, where local Coulomb repulsion opens the coil and reduces the scattering form-factor dramatically. Polymer melts, on the other hand, scatter strongly at low Q , but current studies are resolution limited in observing long-range, low frequency motions. Real advances here require improved Q resolution as well as energy resolutions extending down to the hundreds of μeV range; this is foreseeable with long wavelength neutron spin-echo on a high-flux source. It is also worth noting that in new fields such as the study of polyelectrolytes, even the phenomenology is unknown at the microscopic level, and a predictable use of greatly enhanced flux will initially be to survey and characterize model systems.

2.2 CONDENSED MATTER PHYSICS

It is interesting to recall that one of the basic neutron spectroscopic techniques used today in physical chemistry is backscattering, which was initially proposed, inter alia, for the study of hyperfine interactions in solids. This cross-fertilization effect resulting from having a centralized, user-oriented facility will certainly lead to further new and unpredictable uses of high resolution spectrometers, but one should bear in mind that the more conventional areas for which the spectrometers were conceived will also continue to flourish. In condensed matter physics, there is a continuous stream of interesting and important physics problems for which neutrons provide essential information. Low dimensional materials, for example, can now be synthesized in a remarkable variety of forms whose structure and dynamics can often be studied only by neutron scattering. Studies of in-plane dynamics of intercalates or of excitations in hexatic phases, and the structure and dynamics of absorbed molecular species in such compounds, will require high resolution and a copious flux of cold neutrons. Phonon driven structural phase transformations in these and other materials will also be better understood when the mode softening can be studied at very low frequencies and small wavevectors near the phase transition.

Spin glasses have been the subject of intense experimental and theoretical efforts over the past few years, but many fundamental questions remain unresolved. In these systems, there is a fairly well-defined temperature below which the spins are apparently frozen into random orientations without long-range correlations. A typical example of spin

glass behavior is provided by the Cu-Mn alloys, for which neutron polarization analysis diffuse scattering data show the coexistence of two types of spin correlation: a long period spin modulation and a net ferromagnetic correlation which is directly associated with atomic short-range order. The latter correlations have been studied over a wide range of energy resolutions, and the results interpreted as indicative of a broad distribution of relaxation times with no phase transition to the spin glass state. However, recent studies of the elastic component of the diffuse peak corresponding to the long period modulation show a temperature dependence closely resembling that expected for an order parameter approaching a critical point. Whether this really corresponds to a phase transition cannot be decided at currently available resolutions, and the problem of reducing the present lower limits of relaxation rate awaits higher resolution spectrometers. Here, as in many similar areas, the long relaxation rates are correlated with distances which are large relative to interatomic distances, and so the problems are well suited to study by methods where the increased resolution is obtained by using longer wavelength neutrons.

Another aspect of phase transformation studies is the increasingly pressing need for accurate data with which to compare ab initio calculations of lattice dynamics. Not only frequencies, but also phonon widths can now be calculated quite accurately, and high resolution line-width data (measured, for example, by neutron spin-echo triple axis spectroscopy) are presently lacking. This is only one example of a number of areas in which advances in numerical computations (particularly by Monte Carlo or molecular dynamics techniques) are starting to outstrip currently available experimental resolutions with which to test them.

2.3 CHEMICAL SPECTROSCOPY

In chemical spectroscopy, an important feature of neutron scattering is the wide range of time-scales (10^{-6} - 10^{-14} s) and momentum transfers (10^{-3} - 10 Å⁻¹) which may be observed, so that studies may focus on a wide variety of species, ranging from single proton jumps (in, for example, ionic conductors) to the motion of entire complex structures (such as aggregates in solution). The range of momentum transfer available is often determinant in distinguishing between translational and rotational motion and between

different kinds of rotational motion. Further, the vector nature of the momentum transfer allows anisotropic motions, such as diffusion in intercalates, to be followed in specific directions. With increased flux, techniques such as polarization analysis will also become routine, allowing scattering from individual atoms or groups to be distinguished from that due to correlated motions of many atoms or molecules.

Tunneling spectroscopy, or the study of ground-state rotational splittings, is one such example of a major new physico-chemical technique requiring both high resolution and high flux. Extremely detailed information about the local potential in a variety of systems is obtained by this method. The scattering in this case couples to the wavefunction of the whole molecule, or at least that of the local group, and requires exchange of angular momentum with the neutron spin. Selective deuteration is an important tool, since it varies the spin wavefunction compared with the protonated species. Tunneling may be thought of as the interchange of atoms in a group (e.g., H in $-\text{CH}_3$) at temperatures too low for reorientation to occur. The study of the crossover from tunneling to classical reorientation as the temperature is increased is important, for example, in the study of organometallic complexes, where the entire complex may eventually rotate about a symmetry axis. The variation in frequency and lifetime of a mode with temperature has provided detailed information about potentials in bulk materials, but may prove most important in studying isolated molecules such as hydrogen trapped in impurities in metals or bound in a solid gas matrix. Surface areas are of particular interest, where the probe molecule may be subjected to varying neighbour interactions by altering the coverage or by placing it in or on an adsorbed layer (or layers) of a different species. These studies all require very high flux, since the concentrations are low and the small splittings require extremely high resolution. Translational tunneling, for example in hydrogen-bonded molecules, poses even more stringent requirements on flux and resolution, and so far has eluded detection. The study of spin conversion processes in tunneling systems (e.g., NH_4ClO_4) is possible at current resolution, but needs higher flux to obtain spectra faster than the natural decay rate.

The range of fast ionic conductors in which stoichiometric rearrangements and diffusion mechanisms may be studied will be greatly expanded once there is sufficient flux to allow routine study of the incoherent scattering

from ions other than protons: incoherent cross-sections for most ions are nearly two orders of magnitude smaller than that for $^1\text{H}^+$, and are often coupled with strong absorption cross-sections (in Li^+ , Cl^- , Ag^+ , for example). Similar remarks apply to many surface studies, for example, catalysis.

2.4 COLLOIDAL SYSTEMS

Complex solution phases such as colloidal suspensions, micellar solutions, ferrofluids and microemulsions are particularly well-suited to study by neutron spectroscopy. The entire arsenal of contrast matching techniques is available to allow these very complicated systems to be studied in a controlled fashion. Further, polarization analysis may be used to separate coherent from incoherent scattering, and in the case of ferrofluids (either pure, or as media for magnetic bubbles), polarization analysis provides the only means of obtaining microscopic information on magnetic correlations and fluctuations.

These systems are all large on an atomic scale, and very high resolution is needed to study the concomitantly slow motions. Current studies have exposed many features of fundamental interest, particularly in the area of hydrodynamic interactions in concentrated systems, but at present flux and resolution limitations tend to inhibit further rapid progress. An interesting feature of such systems is that, apart from their major intrinsic interest, they provide excellent models for studying fractal phenomena, many-body hydrodynamics and other areas of more global interest. The latter feature has led to vastly expanded theoretical work in this area in recent years, which in turn has allowed quantitative interpretation of scattering data and has encouraged work on even more complex systems. Even a single surfactant in solution may form spherical or cylindrical micelles, lamellar, cubic, hexagonal and reversed hexagonal structures, and liquid crystalline phases, depending on temperature and concentration; the need for very high flux is self evident even for the study of a single system, before considering the more interesting case of the dynamics of absorbed (and perhaps reacting) molecules in, for example, micelles. Other areas where neutron spin echo studies, in particular, have led to new understanding are the critical behavior near demixing phase transitions in non-ionic micelles and the existence of soft hydrodynamic modes in microemulsion droplets. The

latter, in particular, are currently at the limits of present neutron spin echo spectrometers, and the ability to survey even one (typically four component) microemulsion is flux-limited on existing sources. Improved resolution will also be required before experiments can be extended to the interesting supercooled and glassy phases of these materials.

2.5 BIOLOGICAL SYSTEMS

The use of inelastic scattering to study biological materials is a recent development. However, the recent applications of high resolution inelastic scattering to macromolecular systems suggest the possibility of obtaining fundamental thermodynamic parameters associated with biomolecular properties, and of observing alterations in dynamic states that are correlated with functional changes in biological molecules. A change in the dynamics of hexokinase, for example, has been detected when the substrate glucose is bound to the enzyme. Very high resolution at low to intermediate Q is needed for such studies because of the size of the molecules involved, but appropriate resolution should be available in the next generation machines on a new source. Selective deuteration, where possible, will play an important role here in studying correlated motions of marked sub-units.

3. High Resolution Spectrometers

We were able to give detailed quantitative consideration to possible next-generation high resolution spectrometers, thanks to the prior availability of several source documents¹⁻⁵ and to the generosity of the Jülich group, who made available preprints of their current SNQ design studies.^{7,8,10} In this section, we shall for the most part summarize relevant instrumental concepts, and refer the reader to the references for complete design calculations. In a few cases details will be discussed, especially when the committee was able to amplify existing ideas or to see ways of implementing a new idea on an alternative source. The main techniques are discussed in order of increasing resolution.

3.1 TIME-OF-FLIGHT

The general advantages of a pulsed source for time-of-flight have already been discussed in section 1. Beyond the source gain, we shall also consider instrumental gains which would be achieved on either source.

"Marx" Time-of-flight: Rather than direct detection of the scattered beam, a combination Multi-Angle Reflecting Crystal and multidetector is used, with the large sample in a white incident (chopped) beam. Correlations between the sample-analyser-detector path length and the energy reflected by the analyzing crystal at a given angle are used to compensate for sample size effects. With the graphite in near backscattering, the resolution would be of order 10 μev in the quasielastic channel and 100 μev at 10 mev energy transfer. Note that with this (and any other) instrument which places the sample in the full white beam, incoherent background production may pose certain problems. A variation on this theme would be to use a spectrometer like IRIS (see below) with two circles of analysers in planes above and below the sample, reflecting slightly out of backscattering onto detectors also above or below the sample.

Correlation/Time-of-Flight: The idea of correlation chopping is to use a high duty cycle to gain intensity. Usually, the problem under these conditions is that the technique only provides a useful gain for signals higher than the mean level, so that the presence of a large peak in the spectrum obscures information from the smaller peaks. If coarse time-of-flight-techniques are used (on a pulsed source) to select out only the region of interest, the correlation technique may be gainfully used within a window covering just that region. This technique would be particularly apposite on a source generating relatively long pulses.

Bandpass Filter/Time-of-flight: A bandpass filter allows a fixed final energy (only) to be detected, and time-of-flight techniques determine the incident energy of each neutron. Filters could be cold Be-hot Be, Be-BeO, etc. For very small final energy, a combination of mirrors in reflection-transmission would allow variable bandpass and centre to be selected by adjusting the mirror angles to appropriate critical wavelengths. (This could also be used to select incident bandspreads on reactor-based instruments.) Alternatively, a Drabkin undulator could be used.

Bunching Spectrometer⁷: The bunching technique uses moving crystals to achieve time focussing through Doppler shifting of neutron velocities. The gain depends on the bunching time, t_B , during which the Doppler acceleration can be maintained during the time T between neutron pulses. For a neutron velocity v and a relative resolution α , the machine gain relative to a conventional double chopper spectrometer is

$$g_m = 4 t_B v / L \alpha$$

where L is the source-monochromator distance (= monochromator-detector distance). For $\alpha = 1\%$, current Doppler-drive technology leads to $g_m \sim 12$; this should be compared with $g_m \sim 3$ for a multiple crystal time-focussing spectrometer such as IN6 at ILL. This gain would be compounded with the source gain g_s (see section 1) on a new source.

Cool Double Chopper: In cases where good energy resolution is required at moderately high momentum transfers, a double chopper operating on a cool beam ($E_0 \sim 10$ mev, $\lambda \sim 3\text{\AA}$) could provide resolution of order $\Delta E = 40$ μ ev provided the pulse-width of the second chopper is reduced to ~ 5 μ sec. Despite the severe pulse-tailoring this would require on a pulsed source, the gain factor would still be of order 50 relative to a machine like IN5 at ILL.

3.2 BACKSCATTERING

Musical⁸: This is a modified backscattering spectrometer designed to use the time structure of a pulsed source. The spectrometer is similar to IN10 (ILL), but has the single monochromator replaced by a set of crystals placed one behind the other (see Fig. 1). Each crystal has a different lattice spacing and so selects a different part of the inelastic spectrum. Spacings are chosen such that $E_{n+1} - E_n = \Delta E_M$, the energy resolution of one monochromator. The different monochromator-sample distances produce different flight times for each incident energy; these are sorted by usual time-of-flight techniques (see Fig. 2). The machine gain-factor for such a spectrometer is basically the number of crystals, which in practice can be of order 10-12. The different d-spacings may be produced by keeping the monochromators at different temperatures, or by using crystals of the type $\text{Si}_{1-x}\text{Ge}_x$, which have recently been successfully tested.

Fig. 1 The backscattering spectrometer MUSICAL.

Fig. 2 Time-of-flight diagram for MUSICAL.

Iris⁹: This is a pulsed-source backscattering spectrometer combining a crystal analyzer with time-of-flight techniques to analyze the incident energy. The flight paths necessary for an optimized spectrometer at the highest resolution (1 μeV with silicon analysers) are $\approx 300\text{m}$ long. However, when graphite analysers are employed, producing $\Delta E \approx 10\mu\text{eV}$ in a 1 meV window, an optimized spectrometer need only be 35 m long. On the SNS, a non-optimized silicon analyser (1 μeV in a 90 μeV window) will also be used at 35 m. (In the latter case, a fast chopper close to the moderator will be used to reduce the pulse length.) IRIS would have intensity comparable with MUSICAL, but the full intensity gain would only be realized for a spectrum which filled the window; on MUSICAL, the window (typically $< 30 \mu\text{eV}$ wide)

may be tailored to the spectral width. With IRIS, on the other hand, the window may be centered on any chosen inelastic energy transfer.

Focussed time-of-arrival backscattering: This instrument, proposed by H. Mook, is a multi-monochromator reactor-based backscattering spectrometer. The layout is similar to MUSICAL, but each monochromator is mounted on a separate Doppler drive, and these drives are phased relative to one another.

Fig. 3 Time-distance diagram for focussed time-of-arrival backscattering. Note that neutrons may originate at any time. (Compare with Fig. 2.) On the scale of the diagram, all velocities are the same.

The principle of the spectrometer is based on the fact that neutrons from any of the monochromators which arrive at a given time T_n at the detector all have the same incident energy, due to appropriate choice of the Doppler-drive phasing (see Fig. 3). The monochromator d-spacings are chosen so that $E_{n-1} - E_n = \Delta E$, the resolution. The spectrometer thus gains in intensity by the number of crystals, each of which overlays a copy of the spectrum on the detector. All copies are identical (to first order in $\Delta E/E$), but have asymmetric windows (since the central channels are overlaid after shifting by ΔE). For a few crystals, the end channels may simply be ignored, or an asymmetric Doppler drive may be used to compensate for this effect.

The problem of double scattering in the monochromator system must also be overcome. If perfect back scattering is used with the monochromator

crystals directly one behind the other, the Doppler phasing is such that a neutron reflected with final energy E_n from monochromator m will again satisfy a symmetrical reflection condition just as it arrives back at monochromator $m+1$. After this second reflection (with final energy E_{n-1} into the original beam direction but delayed by 2τ), no further reflections will take place. Monochromators slightly offset from perfect backscattering or some other form of beam diversion will be required to avoid this effect.

3.3 NEUTRON SPIN-ECHO

The layout of a typical current spin-echo spectrometer is shown in Fig. 4. The velocity selector produces a beam whose bandspread is a compromise between high intensity (large $\Delta\lambda$) and reasonable Q resolution (small $\Delta\lambda$).

Fig. 4 Layout of a conventional spin-echo spectrometer. The new proposals would use a broad incident wavelength spread and use energy analysis at the detector.

The energy resolution depends on $1/t_m$, where t_m is the maximum Fourier time available; t_m is proportional to the number of precessions in the field H , which depends on the flight-time (and hence on λ), and is inversely proportional to the neutron energy, so that $t_m \propto \lambda^3$. The use of a white beam with final wavelength analysis would therefore have several advantages:

(a) the dynamic range is proportional to $(\lambda_{\max}/\lambda_{\min})^3$;

(b) the Q resolution may be optimized by appropriate choice of the analyzer wavelength discrimination; and

(c) a scan in Fourier time is obtained from the wavelength scan, at fixed field.

Time-of-flight spin-echo¹⁰: On a pulsed-source machine, final wavelength analysis would be by conventional time-of-flight, with a resolution appropriate to the Q resolution desired. The spectrometer would typically gain a factor of 10 over IN11 (ILL) when used at comparable best resolution, with the added advantages of having better Q resolution and of scanning a number of shorter Fourier times simultaneously. Frame overlap problems, however, reduce this gain factor towards unity if a longer maximum wavelength is used to improve the energy resolution.

White-beam spin-echo: On a reactor-based instrument, a white beam could also be used, with the time-of-flight analysis replaced by a bank of analyzer crystals, probably of the synthetic multilayer type. Although this geometry is normally undesirable (for example, on a 3-axis), the very tight collimation imposed on high resolution spin-echo by magnetic homogeneity requirements allows a feasible geometry in this special case. The advantages of a reactor based instrument would be the same as for the time-of-flight machine, with the added feature that wavelengths of, say, 20Å could easily be used, giving an enormous resolution gain; measurements below 1000 pev energy transfer should become possible.

The main technical problem to be solved is the production of suitable white-beam spin-turn devices; this problem is overcome on the time-of-flight instrument by phasing conventional (wavelength dependent) spin-turn coils to each wavelength as it arrives. One possibility is to use dipolar precession fields, which could be made very homogeneous using superconducting coils since the fields are small (~0.1 T). Meissner effect sheets (probably of

Nb) could then be used to produce spin-turns in the homogeneous field, avoiding the usual end-effect problems. The field outside the Meissner sheets must be kept small, and the small residual precession of the spins compensated by a second echo region between the main fields and the sample (see Fig. 5).

Fig. 5. (a) Schematic spin-turn sequence corresponding to Fig. 4. (b) Double-echo sequence using Meissner $\pi/2$ turns; number of precessions in $L_2 \ll L_1$.

3.4 OTHER TECHNIQUES

Two other techniques deserve special mention. The first is the use of ultra-cold neutrons for high resolution, low-Q spectroscopy. This has been successfully applied to certain polymer problems by the Munich group, and provides a useful complementary method to neutron spin-echo, since $S(Q, \omega)$, rather than $F(Q, t)$, is measured.

The second is the use of phase-space transformations, proposed by the Jülich group. As an example, consider the collimation mis-match in a back-scattering spectrometer of the type shown in Fig. 1: tight incoming collimation is followed by large solid-angle analyzers. Since the technique requires small $\Delta\lambda/\lambda$, an appropriately designed moving monochromator (mosaic) crystal may be used to Doppler-shift Bragg-scattered neutrons in a correlated fashion which swaps wavelength spread for divergence (see Fig.6).

Fig. 6. Example of a phase-space transformation from small $\Delta\theta$, large $\Delta\lambda$ to large $\Delta\theta$, small $\Delta\lambda$ to provide optimal beam characteristics for a backscattering spectrometer. The Doppler velocity of the monochromator is v .

Another example is the Doppler-bunching spectrometer, described in ref. 3.

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CHEMICAL SPECTROSCOPY

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I. Introduction

The purpose of chemical spectroscopy with neutrons is to utilize the dependence of neutron scattering cross-sections on isotope and on momentum transfer (which probes the spatial extent of the excitation) to understand fundamental and applied aspects of the dynamics of molecules and fluids. Current interest is concerned with elucidating the relationship between dynamics and the potential governing molecular motions. Examples of such studies are the determination of force fields and of crystals fields responsible for physisorption, chemisorption, transport on surfaces, intercalation, and adsorption by catalytic materials such as zeolites.

Three unique properties of neutron scattering are of significance for this type of work: (1) the use of momentum transfers between $0.1 \text{ \AA}^{-1}$ and $10 \text{ \AA}^{-1}$ to define quantities such as a) as the eigenvectors of molecular vibrations (independently of the eigenfrequencies which are obtainable from IR or Raman spectroscopy), b) the Elastic Incoherent Structure Factors (EISF) for molecular motion, and the characteristics of this motion such as rotational diffusion by small steps, jumps, or continuous reorientation, ion diffusion, and the scaling of molecular diffusional dynamics and validity of equations of motion for transport over atomic distances; (2) the very large incoherent scattering cross-section of hydrogen and the contrast available because of the neutron scattering operator and through isotopic substitution which allows for the separation of scattering, for instance from a substrate and adsorbate, into the two components and identification of cross terms between them; (3) the very high energy

resolution of neutron scattering at low energy transfers, which has revealed, e.g., tunneling phenomena that are a key to determining self-consistent potentials for the crystal fields mentioned above. One particularly important application of higher intensity neutron sources would be the extension of the high resolution capability to higher energy transfers.

In the following discussion we have divided the area of chemical spectroscopy into three energy ranges, which at present sources are principally covered by three classes of spectrometers, as follow: (1) Vibrational spectroscopy, 25 - 500 meV, for which much of the work is done on Be-filter analyzer instruments; (2) Low energy spectroscopy, ≤ 25 meV, for which the prototypical spectrometers are IN4 and IN6 at the ILL; and (3) High resolution spectroscopy, ≤ 1 meV, which typically is performed on the backscattering spectrometers IN13, IN10, or the TOF spectrometer IN5.

II. Vibrational Spectroscopy

We have identified three major areas for significant improvements on the current work in neutron vibrational spectroscopy that would uniquely be provided by an advanced neutron source. These are the measurement of the Q-dependence of the vibrational spectra, higher energy resolution as well as an extension of the Q-range to much lower values at high energy transfers, and the provision of higher sensitivities. We will now discuss representative examples in each of these categories.

(1) High energy resolution studies which include Q-dependence. Much of the current work involving inelastic incoherent neutron scattering (IINS) vibrational spectroscopy suffers from the fact that data are collected at high momentum transfers (as much as $15 \text{ \AA}^{-1}$). The reason for this is that at many present sources high count rate analyzers must be used to collect this data. These are usually low energy band pass filters for which $|k_f|$ is small compared with $|k_i|$, such that the momentum transfer increases with incident energy. As a result the spectra at high energy transfers are often contaminated with combination and overtone bands and therefore single phonon peaks and their intensities are often difficult to

Figure 1: The IINS spectra of $\text{HCCO}_3(\text{CO})_9$ obtained on the FDS at Los Alamos (top, $T = 15\text{K}$) and the IRS at Harwell (bottom, $T = 90\text{K}$). Note the lack of signal in the C-H_1 stretching region in the FDS spectrum ($Q \sim 13 \text{ \AA}^{-1}$ at 3000 cm^{-1}).

obtain. There is good reason to believe [1], that many of the published spectra of C-H stretching modes by IINS are doubtful because of this contamination [Fig. 1]. The importance of extracting accurate one-phonon intensities from IINS spectra lies in the unique property of neutron scattering, namely that intensities can be calculated exactly and can be utilized for making mode assignments. This procedure then leads to the deduction of force fields, which for instance are needed as input parameters to the calculations of IR intensities, a subject of great importance in spectroscopy.

In order to obtain more accurate one-phonon intensities vibrational spectra must be taken as a function of momentum transfer and with better energy resolution than is currently possible. This will minimize multi-phonon contributions and allow distinction between overtones and single phonon excitations. Such measurements would require, for example, a resolution of at least 2 meV at an energy transfer of 200 meV and a minimum attainable momentum transfer of approximately $2 \text{ \AA}^{-1}$. The instrument of

choice for this type of measurement would be a chopper spectrometer at an advanced pulsed neutron source utilizing short pulse widths with incident energies between .5 and 2 eV and a flux at the sample of at least 10^5 n/cm² sec.

The ability to measure vibrational spectra with much higher resolution is of particular importance for complex molecules, which typically also have the most uncertainties in the assignments of vibrational frequencies. Even prototypical hydrogen-bonded systems (which have been studied extensively by many techniques) are often poorly understood. One such example are the Bifluoride and related systems, where a recent measurement with higher resolution revealed an unexpected splitting in the intense bending mode [2]. This work needs to be repeated with a highly dilute (factor of 100) system to resolve the question on the origin of this splitting. This is just one of many cases of even relatively simple compounds where high resolution is essential for mapping out details of the potential surfaces in question. Furthermore, it would be of great interest to use a single crystal in this case to orient the potential with respect to the incident neutron beam. This would provide additional information on the anisotropy of the potential. It may be desirable to also have a spectrometer with a large focusing crystal monochromator for this type of measurement.

Considerable effort has gone into computing and mapping out of potential surfaces of hydrogen trapped in metals which is an essential step towards understanding the bonding and diffusion processes in these important materials. Most of the experimental work that included the observation of more than the lowest bound states has been limited either to concentrated samples or to measurements without Q-dependence. The potential of dilute H in bcc metals is known to be very anharmonic and the observed transition lines have as yet unexplained line widths. A determination of the full $S(Q, \omega)$ would -- in conjunction with theoretical modelling -- provide detailed information on the proton wave function and the potential. In addition, the transition to recoil scattering of the proton can be studied if a high enough incident energy is available. The range of $S(Q, \omega)$ that would be required for this class of experiments is roughly $\omega < 1000$ meV, and $2 \text{ \AA}^{-1} < Q < 25 \text{ \AA}^{-1}$.

There are numerous other cases where present source intensities do not allow resolution improvements necessary to do the experiments. Recent theoretical work by M. Warner and collaborators [3] has, for instance, pointed out the possibility of studying the coupling between internal and

external modes by measuring the fine structure of the vibrational transitions. Other examples include the exciting possibility of distinguishing either molecules adsorbed on different sites of a catalyst or different molecular species resulting from a catalytic reaction by differences in their respective vibrational spectra, or an accurate determination of the crystal field splittings, e.g., of the rotational transition lines of H_2 in graphite intercalation compounds [4]. The intercalated graphite substrate imposes a relatively weak crystal field on the rotations of the hydrogen molecule which split the transition levels [Fig. 2] by 1-2 meV at energy transfers going above 200 meV. This is a dilute system which requires high energy resolution and low Q to obtain detailed information on the interaction of H_2 with the substrate. Even with the next generation source the energy resolution is such that the data are not likely to be fully resolved. Some curve fitting is thus likely to be necessary. This problem

Figure 2: Calculated crystal field levels of H_2 in a crystal field. Note that for small values of V high resolution is required at large energy transfers to resolve the levels.

is made even more severe because the required optimum Q values ($\sim 1.5 \text{ \AA}^{-1}$) impose such a high E_i that ΔE becomes unacceptably large.

A large number of important spectroscopic experiments need to be done on loosely bound molecules. This has in general not been possible because of the requirement to keep Q well below $3 \text{ \AA}^{-1}$ at as high an energy transfer as possible even if modest energy resolution is acceptable. This would allow the study of physisorbed molecules or molecules in aqueous as well as nonaqueous solution. This has been shown in at least two recent examples where the normal modes of species in concentrated solution have been observed. The first of these [5] involves the observation of the torsional modes of the $\text{N}(\text{CH}_3)_4^+$ ion in D_2O at energies less than 40 meV, while in the second experiment detailed data were obtained [6] up to 450 meV for chloroform (CHCl_3) in liquid sulfur dioxide [Fig. 3]. This demonstrated ability to study species in various types of solution is chemically very important. The method can, however, only be more fully exploited if dilute solutions can be examined and data collected over a wide range of Q , including low Q ($< 6 \text{ \AA}^{-1}$ at 400 meV).

Figure 3: IINS spectrum of CHCl_3 in liquid SO_2

(2) Medium resolution vibrational spectroscopy. There exists a range of problems for which even at an advanced source the intensity will not be sufficient for Q dependent and high energy resolution measurements to be feasible. Despite this the reasons for doing this type of experiment

are essentially identical to those outlined in the previous section.

The factors which contribute to the requirement for such a high sensitivity spectrometer may be listed in terms of the approximate increase in flux at the sample (incl. energy analysis) that is necessary over current spectrometers as follows:

- effect of dilution: 10-100
- smaller cross section samples: 1- 40
- differential spectroscopy: 10

Thus the combined need for intensity increases ranges from a factor of 100 to 40,000! We will now describe some representative examples for each of these categories.

(A) Differential Spectroscopy. Excellent statistics are required if differences at the level of less than a percent are to be observed between samples of very similar cross-sections. These difficulties are aggravated for difference studies on dilute media. Examples include:

- (a) Molecules on the surface of colloidal particles
- (b) Species adsorbed on catalysts, particular supported catalysts since in these cases the surface areas and adsorption capacities are extremely low; this type of work should also be extended to include adsorbates with low cross-section, i.e., molecules not containing hydrogen.
- (c) Molecules intercalated between graphite layers. A great deal of pioneering work has been performed on the dynamics of small molecules on high surface area ($10 \text{ m}^2/\text{gm}$) graphite substrates. At energy transfers below 50 meV, for example, the vibrational density of states of adsorbed CH_4 shows peaks

Figure 4: Vibrational spectra of 0.7 monolayers of methane adsorbed on exfoliated graphite, $T = 10\text{K}$, with Q parallel (a) and perpendicular (b) to the surface (Ref. 7).

[7] resulting from motion perpendicular and parallel to the surface as well as librational excitations in the crystal field that the surface provides [Fig. 4]. This type of measurement can be done with an energy resolution of ~ 1 -3 meV, i.e., rather better than is possible with the best alternative techniques, e.g., electron energy loss spectroscopy. An increase in neutron source flux would therefore allow spectra of this quality to be taken -- even with current spectrometers -- on a much wider range of substrates whose specific surface area is much less than that of graphite.

- (d) Modulation experiments. An example of this category would be the study of changes that take place in an electrode surface layer as a function of changes in the applied potential.
- (e) Isotopic substitution. The most common case is the selective replacement of hydrogen with deuterium because of their large difference in cross-section. This is an extremely powerful aid in the assignment of vibrational spectra. Higher neutron source fluxes would allow other substitutions, such as $^{14}\text{N}/^{15}\text{N}$ or $^{35}\text{Cl}/^{37}\text{Cl}$. For example, it would be desirable to study the DF_2^- ion at ca 1% incorporation in an alkali halide lattice in order to help answer questions (see Section II.1) on this basic hydrogen bonded system. The bifluoride cross-section at this dilution would be reduced by a factor of 1000 over that of the parent NaHF_2 .

(B) Time-dependent processes. For compatibility with other techniques it is desirable to obtain spectra in very short times. By reasonable extrapolation of current spectrometer performance it can be shown that in favorable cases a vibrational spectrum can be taken in about one second at an advanced neutron source. This makes it feasible to study a range of relatively slow, but nevertheless important chemical phenomena. These include, for example:

- (a) Reactions in alumino silicate frameworks, e.g., the isomerization of cyclopropane to propene, described below;
- (b) The competitive adsorption of reactants and products or the changes which occur during the displacement of one species by another;
- (c) Changes which accompany intercalation and adsorption.

During the isomerization of cyclopropane over some zeolites (e.g., ZnNaA)

it has been shown [8] that the production of propene can be followed over a time period from one second to one hour. During this period the relative intensities and frequencies of the IR bands from the adsorbed reactant change significantly. Since the zeolite is strongly absorbing below 150 meV, several of the internal modes cannot be observed using IR. Neutron scattering studies would make it possible to supply the missing data as well as to give information on the important modes of the zeolite/ C_3H_6 system, i.e., those which directly reflect the strength and nature of the interaction of the adsorbed molecule with the surface.

For the above types of measurements a spectrometer with very high sensitivity is required. This suggests an instrument which integrates counts over a large solid angle (no Q resolution), an advanced analog to such instruments as the FDS (Los Alamos), CAS (Argonne), the INIB (ILL), or the BT-4 (TRASH) at NBS. It is essential, however, that the energy resolution be superior to that available at this time, so that reasonably well resolved spectra can be obtained for mode assignments and intensity calculation. A factor of two improvement in resolution could then be combined with the other factors mentioned at the beginning of Section II.2 to imply a required intensity increase of a factor 400 to more than 100,000 over current instruments at medium flux sources for this type of spectroscopy.

III. Low Energy Spectroscopy ($E < 25$ meV)

The future needs for chemical applications in this energy regime lie in at least three different areas, two of which are analogous to the case of vibrational spectroscopy, namely better energy resolution on the one hand, or higher sensitivity on the other. A third, potentially very important class of experiments would utilize polarization analysis.

(1) High energy resolution studies. The first class of experiments requires the attainment of even better energy resolution (10-20 ueV) in order to allow for example the probing of molecular motions which more closely overlap the NMR range. This must be achieved with the same high sensitivity as discussed below and over a Q range extending to $2.5 \text{ \AA}^{-1}$ in order to unravel the geometry of long-range diffusive and rotational motions. A large number of systems involving isolated or weakly interacting molecular groups could be investigated with such a measurement capability. For example [9], existing facilities at the ILL and at NBS have been used to establish for the first time the detailed potential hindering the rotation of the methyl group in the solid phase of nitromethane [Figs.

Figure 5: Inelastic neutron scattering spectra of CH_3NO_2 at 4.2 K and pressures of 1 bar and 4.8 kbar. The top part shows the pressure dependence of the ground state tunnel splitting, the bottom part that of the first excited torsional level.

5 and 6]. However, it is not at present possible to extend these measurements to dilute molecular systems.

The ability to probe molecular motion and processes in this range of Q and energy transfer will open up important new research opportunities in many chemical systems, including transport mechanisms in glassy or crystalline superionic conductors, chemical exchange reactions, solvent exchange in colloidal suspensions, and the dynamics of coordinated molecules around complexes in ionic solutions, as well as on chemisorbed or intercalated molecular species. Such instruments would have a great impact on similar studies which are at the interface between physics and chemistry. A recent example is the unique study of hydrogen tunneling around chemical impurities in superconducting metals at extreme dilution. The effects of a

Figure 6: The rotational potential of nitromethane as a function of methyl group rotation with parameters corresponding to 1 bar.

Figure 7: Tunneling states of H trapped at oxygen impurities in Nb. The sample was 50 g of NbO_{x-y} with $x = y = 0.00015$.

magnetic field on these tunneling states in studies of oxygen traps in Nb on the IN6 spectrometer at the ILL are shown in Fig. 7.

(2) High sensitivity studies. For a second class of problems it would be desirable to achieve still higher sensitivity using neutron beams with incident energies of 10-20 meV and Q 's up to $5 \text{ \AA}^{-1}$ at a resolution of about 0.1 meV with a large solid angle of detection. This capability is essential to allow, e.g., the measurement of hindered rotational or torsional levels in dilute molecular species bound in chemical adsorbents, catalysts or in layered or intercalated materials, as well as for the study of the form factor or diffusion paths of species undergoing reorientation or diffusion. Such information is the key to a fundamental understanding of bonding potentials and local transport of chemical fragments or species in dilute media, and to probe the effect of intermolecular interactions on these potentials. These interactions are often a dominant factor in determining reactivity and exchange in chemical systems.

Figure 8: Density of states of additional low frequency modes in vitreous silica compared to the Debye density of states (Ref. 10).

Such an instrument would also permit a wide range of other applications, such as studies [10] of low energy excitations in disordered materials [Fig. 8] which are critical to understanding the low temperature specific heat anomalies in these systems (fractal behavior, two-level systems, etc.). The search for low frequency motions of macromolecules, which are postulated to be a key to the biological activity of enzymes, is

another area that requires extremely high sensitivity since those typically are difference experiments on very small samples looking for relative changes of presumably less than 1%. Neutrons have a unique role to play in those areas, and the increased flux required to do this experiments (at least 10^5 n/cm²-sec on the sample including energy analysis) is crucial.

(3) Polarization analysis. The quasielastic region, however, contains not only incoherent scattering from the dynamical process of interest but also coherent Bragg scattering, especially when the shorter incident wave lengths are used, which is particularly severe in the case of adsorbed molecules. Measurements of the Q dependence of intensities of the quasielastic scattering in these cases have therefore proved nearly impossible. A possible answer to this interference is the use of polarization analysis. Preliminary considerations show this to be technically feasible with the higher flux of an advanced neutron source. The idea was demonstrated fifteen years ago by Moon, Riste, and Koehler, but has not been extensively applied for lack of efficient, cheap, and simple neutron polarizers and analyzers.

The method depends on the fact that the neutron nuclear interaction is spin-dependent of the form

$$V(r) \propto \underline{S} \cdot \underline{I} \delta(\vec{R} - \vec{r}_I)$$

where S and I are the neutron and nuclear spin angular momenta, respectively. The angular momentum operator can be expanded into $S_{\pm}I_{\pm}$, $S_{+}I_{-}$, and $S_{-}I_{+}$, where the first term is associated with coherent, non spin-flip scattering and the other two with incoherent scattering processes. Thus for an incident polarized beam an analyzer could select that part of the scattering which has conserved spin angular momentum and is coherent. In the example of methane chemisorbed on graphite this would contain the Bragg scattering of the substrate ($I = 0$ for ¹²C) and the coherent part of the methane structure factor. The incoherent part coming from translational diffusion, for example, would appear in the spin flip cross section.

The experimental possibility of such a separation has recently been demonstrated (with marginal statistics) on the new D7 instrument at the ILL by Prandl and collaborators in a study of the reorientational motions of C₂Cl₆ in its plastic crystalline phase [11]. Much higher intensity is clearly required here to perform such measurements on a more routine basis.

Many chemical problems would benefit from the installation of polarization analysis capability in the millivolt to microvolt energy transfer

regimes. Among these are: rotational diffusion processes in plastic and liquid crystals; oscillations and diffusion of physisorbed and chemisorbed molecules; measurements of the density of states in glasses and polymers at low energy transfers; solvent and solute motions in ionic and other solutions.

IV. Very High Resolution Spectroscopy

When a rotating molecule, such as one physisorbed on graphite, is cooled down, the rotational diffuse motions probed in the lower energy resolution measurements mentioned above slow down as jumps over barriers provided by the surface potential become less probable. For molecules with small moments of inertia (e.g., H_2 , CH_4 , etc.) rotational tunneling through

Figure 9: Rotational tunneling spectra ($T = 4K$) of 0.7 monolayers of methane adsorbed on exfoliated graphite. (a) Assignments and polarizations of the transitions; (b) Q parallel to the surface; (c) Q perpendicular to the surface.

a barrier may then be observed at low temperatures as an inelastic fine structure near $E = 0$ [12]. The study of tunneling spectra in molecular crystals is now well established and has also been utilized for both adsorbed H_2 and CH_4 [Fig. 9]. Complete interpretation of the positions intensities as well as polarization vectors leads to unique and extremely detailed information on the crystal field at the site of the tunneling molecule [13]. The application of these methods to a wide variety of molecules in dilute systems or adsorbed on a range of substrates could follow from the availability of higher intensity sources and neutron polarization analysis as discussed above (Section III.3). Other extensive areas of great importance in chemistry that would become accessible to a high resolution ($\sim 1-10 \mu\text{eV}$ for $0.3 \mu\text{eV} < \Delta E < 5 \text{ meV}$) instrument with a Q range up to $2.5 \text{ \AA}^{-1}$ are those where kinetic or exchange phenomena are involved. These include, for example:

- (a) Exchange processes -- Between sites in solids
 - Between ions and solvents in solution (coordination spheres)
 - Around the surface of colloidal and polyelectrolyte particles
- (b) Diffusion
 - In molecular liquids
 - In superionic crystals
 - Of hydrogen in metals

Such a spectrometer is also required to extend the current work on the dynamics of macromolecules. Some of the above topics are also covered in more detail by the report of the subcommittee on Ultra High Resolution Studies.

V. Concluding Remarks

It should be apparent from the above discussion that the scientific opportunities in the area of chemical spectroscopy at a high intensity neutron source are enormous. Furthermore, we have identified several areas (such as time-dependent measurements) which are threshold experiments for a factor of ten increase in source flux and would not be possible by parallel improvements in instrumentation alone.

We have not considered questions on the types of spectrometers or pulsed vs. steady state neutron source in any detail. Available calculations [14] suggest that with one exception the experiments described in the body of this report could be equally well (to within factors of two or so)

be performed on appropriate pulsed source or reactor instruments. The exception to this are experiments requiring high energy transfers with a wide range of Q (Section II.1), which can only be done on a "true" (i.e., short pulse) pulsed neutron source because of the very high incident energies that are necessary.

Most of the examples identified as "high sensitivity" studies would require a flux on the sample of at least $10^5 \text{ n/cm}^2 \cdot \text{sec}$ (including energy analysis after the sample in the case of inverted geometry spectrometers). We have therefore included [Fig. 10] some rough calculation of what resolution could be achieved at a 10^{17} peak flux pulsed source with a chopper spectrometer given the above requirement for flux on the sample under the condition $E_f = 1/2 E_i$. This figure shows that most of the resolution/flux requirements noted in our discussions could be met at an advanced neutron

Figure 10: Calculated time-averaged neutron flux on the sample for chopper spectrometers at an advanced pulsed neutron source (peak flux $10^{17} \text{ n/cm}^2 \cdot \text{sec}$) for a given energy resolution and incident neutron energy under the condition $E_f = 0.5 E_i$.

source. It should also be pointed out, however, that this figure does not address the question of Q-resolution nor does it include polarization analysis.

We can summarize the minimum spectrometer needs at an advanced neutron source for chemical spectroscopy as follows:

I. Vibrational Spectroscopy ($25 \text{ meV} < E < 500 \text{ meV}$)\

- a) High resolution/Wide Q-range: $\Delta E \sim 1 \text{ meV}$, $1 \text{ \AA}^{-1} < Q < 10 \text{ \AA}^{-1}$
- b) High sensitivity: $\Delta E \sim 2\text{-}3 \text{ meV}$

II. Low Energy Spectroscopy ($E < 25 \text{ meV}$)

- a) High resolution: $0.01 \leq \Delta E \leq 0.3 \text{ meV}$
- b) High sensitivity: $\Delta E \leq .5 \text{ meV}$ $Q \lesssim 5 \text{ \AA}^{-1}$;

III. Ultra-high resolution spectroscopy ($E \lesssim 2 \text{ meV}$)

- $0.001 \leq \Delta E \leq 0.002 \text{ meV}$, $Q \lesssim 2.5 \text{ \AA}^{-1}$ incl. polarization analysis

It may, of course, be desirable to cover each of these five basic instrumental ranges with more than one, better optimized spectrometer.

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HIGH ENERGY EXCITATIONS

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I. Introduction

This section addresses the opportunities for the neutron scattering investigation of phenomena characterized as high energy excitations. This area is one of the few considered at this meeting which is a new domain of research rather than merely an extension of areas already explored. The new pulsed neutron sources, which provide very high effective fluxes of neutrons in the electron-volt energy range make this area ripe for exploration and exploitation.

For the purposes of this discussion, high energy excitations are defined as those in which the excitation energy (ω) is greater than 100 meV, that is, the energy exchange (ϵ) experienced by the neutrons upon scattering is greater than 100 meV. Since these states are not normally populated at room temperature, their study can require incident neutrons with energies (E_0) much higher than in the thermal range. A realistic upper bound for E_0 has been set as 10 eV. Kinematic constraints can also force the use of large incident neutron energies even though the excitation energy (ω) may be less than 100 meV if simultaneously the momentum change (Q) must be small.

The study of high energy excitations of neutron scattering has been difficult.¹ The lower boundary of this range has been crossed marginally with existing neutron sources with only a few exploratory ventures into its heartland. The difficulties are that the fluxes of neutrons of the required energy have been low and that the resolution of ω and Q are only obtained at a high price in counting rate. However, this situation should improve as new neutron scattering instruments and sources adapted to this range come into operation.

One advantage of neutrons as a probe of condensed matter when compared to other forms of radiation is that with neutron scattering one can select both

the ω and Q of the excitation to be studied. For $\omega > 100$ meV there are two ranges of Q that are of primary interest. The first is the small Q range, that is $Q < 4 \text{ \AA}^{-1}$ (and in some cases $Q < 1 \text{ \AA}^{-1}$). For example, only at small Q 's are form factors large enough to observe significant count rates for magnetic excitations. The second is the large Q range, that is, $Q > 30 \text{ \AA}^{-1}$. In this range the effects of final state interactions should be unimportant and the impulse approximation holds providing for the experimental determination of momentum distributions.

This report highlights four general areas of investigation which have exciting possibilities for study by neutrons. These are: (1) magnetic excitations in solids; (2) momentum distributions of solids and quantum liquids, especially liquid helium; (3) vibrational excitations of light atoms in solids; and (4) excitations in liquids, gases and amorphous solids. The observation of electronic excitations will require neutron fluxes beyond those that will be produced by the bench-mark neutron sources. In-beam experiments, while not exactly conventional neutron scattering, are briefly mentioned as holding interesting scientific potential.

The committee has evaluated the bench-mark neutron sources for high energy excitation experiments. In general, it is concluded that the pulsed sources deliver prepared neutron beam currents 10 to 10,000 times more intense than the steady state reactors even with hot sources; therefore, only the pulsed sources will meet the needs for the identified experiments. The basis of this conclusion is discussed in Section III.

Development of new instruments that can realize good energy resolution and good statistical accuracy will be critical to progress in measuring high energy excitations. A few types of instruments have been tried and these are discussed. They are: (1) phased choppers, (2) resonance instruments, (3) spectrometers with crystal analyzers and (4) band pass filter analyzers. A number of yet untried types of instruments have been suggested and these are listed.

II. Science

The advent of intense sources of high energy neutrons from new pulsed spallation sources and reactor hot sources has provided the opportunity to extend scattering function ($S(Q, \omega)$) measurements to new regions of Q - ω space which cannot be reached by conventional instruments using low energy neutrons. Figure 1 shows this new domain for neutron scattering involving E_0 's up to

10 eV as compared to the traditional reactor domain up to 100 meV. One can see that high energy neutrons open up: (1) measurements at higher energy transfer ranges (area a); (2) measurements at lower Q values at modest energy transfers (area b); and (3) very high momentum transfer experiments (area c). In the following sections the committee has outlined many of the new scientific opportunities with high energy neutron spectroscopy that will be made accessible by the new high flux neutron sources.

A. Magnetic Excitations

From many investigations of magnetic excitations in the last two decades in the thermal and cold region, it became clear that in most systems the magnetic response functions have either long tails or additional inelastic intensity extending far into the high energy region. A complete understanding of magnetic phenomena requires the knowledge of the whole magnetic excitation spectrum on an absolute intensity scale. The existence of the high energy part in the magnetic response is due to the fact that magnetism in condensed matter is produced by unpaired electrons. Strong exchange interactions, as for example in 3d-systems, lead to steep collective excitations (magnons) merging into a broad particle-hole continuum (Stoner excitations). Stable moments in f-electron systems, on the other hand, show a sharp single particle excitation spectrum (crystal-field and spin-orbit splitting) ranging from μeV to eV, modified by interactions between different ions in concentrated systems. Stable moments become unstable through a strong hybridization of local (f) and delocalized (conduction) electrons exhibiting a broad and far reaching magnetic response (Kondo effect, intermediate valence phenomena, heavy Fermions).

New experiments to start with include the measurements of (1) optic modes of spin waves of iron below T_C ; (2) spin fluctuations in Fe and Ni above T_C ; (3) crystal field levels and magnetic responses in rare earths and actinides; (4) broad excitation spectra in mixed valence and Kondo systems; and (5) perhaps more difficult, Stoner excitations in 3d metals.

An example of new science made possible with epithermal neutrons is the measurement of the crystal field excitation in BaPrO_3 at 260 meV measured at IPNS, (see Figure 2). From the experience of this kind on existing pulsed spallation sources, the subcommittee estimates that an upper limit of 1 eV energy transfer at around $Q = 2$ to $5 \text{ \AA}^{-1}$ is achievable in inelastic magnetic neutron scattering using an advanced pulsed spallation source.

B. Momentum Distributions

By scattering with high energy neutrons at large values of Q where the recoil energy is well above the kinetic and potential energies of the system, one approaches the impulse approximation limit where the scattering law is determined by the momentum distribution $n(p)$. Of interest are momentum distributions in solids and quantum fluids and impurities in solids. Experiments to date at existing pulsed sources have aimed at the kinetic energy distribution in solid ^4He , the Bose condensate fraction $N_0(T)$ in superfluid ^4He , momentum distributions in pyrolytic graphite for comparison with theoretical fits to vibrational phonon spectroscopy, hydrogen in metals and hydrogen in the very anharmonic potentials in hydrogen bonds. Much of this work requires systematic studies as a function of external variables such as pressure and temperature, much better resolution to approach the intrinsic broadening due to final state interactions, and higher Q values to minimize corrections. In particular, while the recoil energy varies as Q^2 , the width varies as Q , so that improved resolution at higher energy transfers is a necessity. Thus, this area would benefit enormously from an advanced pulsed spallation source.

Quantum solids and liquids are a testing ground for many-body theories. Areas for future research in quantum liquids include the systematic study of the Bose condensate in ^4He , the exceedingly difficult measurement of the Fermi distribution in ^3He and in ^3He - ^4He mixtures, and the study of monatomic spin polarized hydrogen. These have been the subject of considerable theoretical work which could be tested by these experiments.

The study of hydrogen in metals and hydrogen bonds is complementary to other techniques for determining the potentials. Vibrational spectroscopy, diffraction and impulse-approximation scattering are sensitive to different aspects of the hydrogen potential. For example, diffraction from double well potentials cannot distinguish between static and dynamic disorder, while impulse approximation scattering can. It would be of particular interest, for example, to study hydrogen bonded ferroelectrics as the systems pass through phase transitions.

Studies of pyrolytic graphite^{2,3} have revealed large discrepancies between model fits to the low energy phonon dispersion curves and observed momentum distributions. Again, let it be noted that spectroscopy and momentum distribution studies are complementary.

At the present time, there is a controversy as to which theories best

explain final state interaction effects. This controversy exists because until now experiments haven't been able to reach the range in which these theories disagree. It is of fundamental interest to conduct very high resolution experiments which would sort out this controversy.

C. Vibrational Spectroscopy Including Local Modes of Light Atoms in Solids

In the field of chemical spectroscopy (see area B in Figure 1; also for specific considerations see the separate subcommittee report on this subject) competition with optical spectroscopy methods is significant, but neutrons are advantageous in many cases because: (1) neutrons interact by different selection rules, (2) special atoms can be labelled by appropriate D/H substitution, (3) in situ experiments can be performed because neutrons see through even complicated sample containers or catalyst supporting materials and (4) neutrons sense the ϵ and Q dependence simultaneously.

Furthermore, inelastic neutron scattering is a unique tool in the investigation of local vibrations of H in metals (and other materials opaque to photons) because of the high incoherent cross section of H for neutrons. The typical energy transfers for molecular vibrations or H-oscillations in metals are below 0.5 eV (H-H vibrations). In some cases, where the determination of higher harmonics is of interest, higher energy transfers may be desirable. The form factors $Q^2 e^{-\langle u^2 \rangle Q^2}$ of these vibrations are maximal for Q -values between 5 and 8 $\text{\AA}^{-1}$. Thus, in general, large energy transfers together with large momentum transfers are suitable. For special and interesting cases like line width measurements, where multiphonon contributions are important, also small Q , high energy transfer measurements are necessary, demanding high incident energies. The combination of high energy diffraction, momentum distributions and spectroscopy will give a detailed understanding of the potential seen by H (and other light atoms) in solids.

D. Liquids and Amorphous Materials

The Q - ω range for experiments to investigate long range correlated motions (see Figure 1) in liquids and amorphous solids (acoustic wave like modes and possible fractal excitations) generally requires low angle scattering and high incident energies to achieve even these modest energy transfers at low Q values. Such a range, extending to higher energy transfers, is also appropriate to the measurement of spin waves in amorphous alloys and optic modes in molten salts (see Figure 1). Complete determinations of $S(Q, \omega)$ in a still wider range of Q - ω (see Figure 1) would allow

tests of Placzek corrections applied to defraction measurements of $S(Q)$. Energy transfer resolution requirements are modest, ~10%.

E. Neutron Induced Excitation Detection (NIED)

High energy neutrons can create excitations in condensed matter which can be directly detected. These include e-h pairs in semiconductors, luminescence, and dissociation products in neutron induced chemical reactions. Such experiments could use the TOF of the neutron to determine the incident energy, and there may be coincidence experiments where the scattered neutrons would be detected along with the signature of some other excitation so that the total momentum and energy transferred would be known. This group of experiments is called in-beam experiments in this write up.

One example of an in-beam experiment is the use of neutrons to create excitations (e-h pairs) across the band gap in semiconductor devices. An avalanche device in Si can have up to 90% efficiency for detecting e-h pairs. Such an experiment would measure the total cross section and would avoid the problem of detecting a low angle scattered neutron. The results would be complementary to optical probes and should reveal the usual band edge Van Hove singularities. For example, estimates for WNR/PSR and SNS sources suggest 50 counts/sec for such experiments, well above the dark currents.

Another example would be using a neutron to induce motion along the potential energy curves of a molecule. If the ground potential curve crosses another potential curve, it is possible to make transitions to other electronic states which might decay by luminescence or by unimolecular reactions. Unlike other probes, the neutron can impart significant momentum and, by isotope substitution, particular atoms in molecules can be selected. Thus, the neutron could be highly complementary to other methods of molecular spectroscopy. Again, by appropriate choice of optical instrumentation, the sensitivity of the excitations created can be extremely high. A neutron might also induce a highly excited molecule which could then be studied by other techniques such as optical spectroscopy.

While these applications are highly speculative, the advent of the high epithermal flux of new pulsed sources should make in-beam experiments feasible. Such experiments would use very unconventional instrumentation, which highlights the need for general purpose test beam lines at the new facilities.

F. Problems

In this section on "Science" some general experimental problems that are encountered when doing measurements of high energy excitations are outlined. The extension to measurements at high ω and high Q does not impose significant problems related to available solid angle and collimation; however, significant improvements have to be realized for improved energy resolution and counting statistics. The latter is related to counter efficiencies. Of a more fundamental nature are the problems arising for experiments at high energy transfer (0.5 eV) but moderate or low Q ($< 5 \text{ \AA}^{-1}$) (i.e. magnetic excitations which are limited by the form factor) and experiments at moderate energy transfer (50 meV) but rather low Q ($0.5 \text{ \AA}^{-1}$). In both cases the kinematical conditions of the scattering process require large incident neutron energies ($E_0 > 0.12(\hbar\omega/Q)^2$ where E_0 is in meV, $\hbar\omega$ in meV and Q in $\text{\AA}^{-1}$) and scattering angles of only a few degrees. This will reduce considerably the available solid angle and a rough calculation of the expected counting rate $\dot{c}$ in the detector yields the relation;

$$\dot{c} \propto (E_0^{-3} \text{ or } k_0^{-6} \text{ or } (Q/\hbar\omega)^6) * (dE)^2 (dQ_{\perp})^4 * e$$

where dE is the energy resolution, $dQ_{\perp}$ is the transverse component of the momentum resolution, and e the counter efficiency. This strong dependence of the counting rate on the desired energy and momentum transfer makes any significant increase in $\hbar\omega$ or decrease in Q immediately into a severe intensity problem, unless relaxed momentum and energy resolutions may be tolerated. It can be hoped that focusing methods will be devised for special classes of problems. At the same time the sample induced background increases with increasing incident neutron energies (i.e. see Buyers and Loewenhaupt in the HEECM Meeting¹). Multiple Bragg scattering increases as k^6 . Also multiple inelastic scattering comes to play a significant role as a source for background in the small angle counters. Thus it is very desirable to optimize the incident energy taking the above mentioned considerations into account.

III. A Generalized Comparison of the State-of-the-Art Pulsed Source with the "5 x 10¹⁵ Reactor" for Spectroscopy at 1 eV

In Figure 3 the flux spectrum from the bench-mark neutron sources are presented. The effectiveness of the state-of-the-art pulsed source can be compared to the $5 \times 10^{15} \text{ n/cm}^2\text{sec}$ reactor, in a rather general way, roughly following the approach put forward some time ago by Collin Windsor⁴. Consider the flux at a position such a distance from the source that a pulsed beam in

each case has a given energy resolution. In the pulsed source case, the source pulse defines the resolution, and in the epithermal range

$$\frac{\Delta E}{E} = \frac{2\Delta t}{E}; \text{ and } \frac{\Delta E}{E} = \frac{2v\Delta t}{L} = \frac{1}{18L}$$

for a typical moderator ($\Delta t = 2\mu\text{sec}/\sqrt{E(\text{eV})}$). In the reactor case, it is assumed that it is possible to provide a chopper with the same open area as the pulsed source moderator ($A = 100 \text{ cm}^2$), that the chopper can see an arbitrarily large area of the reactor source at the specified flux, that the chopper pulse width is $\Delta t_c = 2 \mu\text{s}$ and that its frequency is $f_c = 500 \text{ Hz}$. The chopper is then the point of origin of the pulsed beam from the reactor. The hypothetical chopper is near the limit of technical feasibility and is well suited to spectroscopy with epithermal energies. We ignore the "lighthouse effect" of the chopper resolution, namely the effect on the chopped pulse due to finite collimation. For the pulsed source

$$\bar{\phi}_s(E) = \frac{d^2\bar{\phi}(E)}{d\Omega dE} A \frac{1}{L^2}.$$

Taking

$$1/L = 18\Delta E/E \text{ (m}^{-1}\text{)}$$

$$\bar{\phi}_s(E) = \frac{d^2\bar{\phi}(E)}{d\Omega dE} A (18)^2 (\Delta E/E)^2$$

then for the reactor source

$$\bar{\phi}_s(E) = \frac{d^2\bar{\phi}(E)}{d\Omega dE} A \frac{1}{L^2} f_c \Delta t_c$$

since

$$\Delta E/E = 2v\Delta t_c/L, \quad 1/L = \frac{1}{2v\Delta t_c} \frac{\Delta E}{E}$$

so that

$$\begin{aligned}\bar{\phi}_s(E) &= \frac{d^2\bar{\phi}(E)}{d\Omega dE} A \left(\frac{1}{2v\Delta t_c} \frac{\Delta E}{E} \right)^2 f_c t_c \\ &= \frac{d^2\bar{\phi}(E)}{d\Omega dE} A \left[\frac{f_c \times 5.23 \times 10^{-9}}{4E(\text{eV}) \Delta t_c} \right] \left(\frac{\Delta E}{E} \right)^2\end{aligned}$$

since $E(\text{eV}) = 5.23 \times 10^{-9} v$ (m/sec). Taking $E = 1$ eV for example, one computes

$$\left[\quad \right] = \left[\frac{5.23 \times 10^{-9}}{4 \times 1.0 \times 2 \times 10^{-6}} \right] = 0.33 \text{ m}^{-2}$$

The ratio of quantities in brackets represents the ratio of effectiveness of the pulsed source and the chopped reactor source for spectroscopy at 1 eV.

Extending this analysis in a simple way for different energies (which is

$$f_c \propto \sqrt{E}$$

$$\Delta t_c \propto 1/\sqrt{E}$$

neither optimal nor violently wrong, except that it is unrealistic at highest energies). The flux at the position where $\Delta E/E = 0.01$ has been plotted (see Figure 4). The result, except for a scale factor, is actually independent of $\Delta E/E$.

The data on angular current density and pulse width given by Crawford and by Moon⁵ for the two types of sources, extrapolated rationally to higher energies, has been used. Since the basis of this comparison is very generous to the steady source at high energies, a more realistic curve based on more conceptually realistic choppers is shown. Figure 4 presents the results, which show that, for high energy spectroscopy in general, the pulsed sources provide 1-4 orders of magnitude greater flux in a beam from the pulsed source, than a similar quality beam from the high-flux reactor with hot source and chopper. Because of this 1-4 orders of magnitude, the subcommittee concluded that the pulsed source is superior for experiments to measure high energy excitations and considered only pulsed source instruments.

IV. Instruments for High Energy Spectroscopy

A number of different spectrometer types need to be tried and evaluated for measuring energy changes greater than 100 meV. For many measurements of discreet excitations, observation at small Q is required, which demands E_0 's much greater than 100 meV. For measuring momentum distributions, observations at large Q 's must be observed, which again demands E_0 's much larger than 100 meV. In both cases, very good energy resolution is required. Recently several types of instruments have been tried while a number of proposed types of instruments are awaiting trial. At present the most promising spectrometer types are: (A) chopper spectrometers, (B) resonance instruments, (C) MAX type crystal analyzer spectrometers (TOF + Bragg crystal) and (D) TOF + fixed-low-energy-pass spectrometers.

A. Chopper Spectrometer

In this type spectrometer, a neutron chopper is placed a distance L_0 down the beam line from the pulsed neutron target. This chopper opens to pass neutrons at a definite time after the neutron pulse is produced at the target; thus the chopper passes burst of neutrons of only one energy. These neutrons are scattered from a sample and the energy of the final neutrons is determined by TOF from the sample to detectors placed at angles and flight paths L_f beyond the sample.

In experiments where one wants to measure large ϵ at small Q , $\Delta E_0 \sim \Delta E_f = \Delta E$ where ΔE is energy uncertainty, and the suffixes 0 and f indicate initial and final. Here ΔE is expressed by $\Delta E \sim 2E\Delta t/t = 1.7/\sqrt{E(\text{eV})/36(L(\text{m})/\sqrt{E(\text{eV})})}E = 0.047E/L$ where Δt is the time uncertainty. From the optimum conditions one has $\Delta t_{\text{moderator}} \sim \Delta t_{\text{chopper}}$.

Consider a typical experiment with $\hbar\omega \sim 330$ meV. From the kinematic diagram, $E_0 = 1 - 2$ eV with $\theta < 5^\circ$. If one wants to perform the experiment with $\Delta\hbar\omega/\hbar\omega \sim 0.05$, $\Delta Q/Q \sim 0.1$, and $E_0 = 2$ eV, then

$$\Delta\hbar\omega/\hbar\omega = \frac{\sqrt{2\Delta E}}{0.330} = 0.05$$

and

$$\Delta E = 0.0117.$$

For these conditions $L_0 \sim L_f \sim 0.047E/\Delta E \sim 10$ m.

The actual value of $\Delta t_{\text{chopper}}$ will be limited to about 2 μs and therefore at the higher E_0 (>1 eV) range

$$\Delta t = (\Delta t_{\text{chopper}}^2 + \Delta t_{\text{moderator}}^2)^{1/2} \sim \Delta t_{\text{chopper}} = 2 \mu\text{s}$$

and

$$\Delta E = \frac{2E}{36L/\sqrt{E}} = 0.056E^{3/2}/L.$$

Next consider a crossing point at which the resolution of the chopper spectrometer becomes equal to that of the resonance instrument. Energy resolution of about 50 meV has already been achieved by some resonance foils at low temperature (^{240}Pu ($E_r = 1$ eV), and ^{238}U ($E_r = 6.67$ eV)). Then the crossing point E_C is expressed by

$$E_C \sim \left(\frac{0.05}{0.056} L \right)^{2/3} \sim L^{2/3}.$$

Then $E_C = 4.6$ eV with $L = 10$ m, and $E_C = 7.36$ with $L = 20$ m, for example.

B. Resonance Instrument

In this type of spectrometer the enhanced interactions of neutrons at a strong nuclear resonance as compared to the weak interactions off resonance are used as a way to identify the energy of some neutrons. In one subtype, the strong capture at resonance is used to detect and identify the energy of scattered neutrons, that is, the resonance foil is used as an energy distinctive detector. In a second subtype the strong resonance is used as a beam filter to remove neutrons of a distinctive energy from the incident or scattered beams of neutrons.

The resonance detector spectrometer (RDS)⁶, the resonance filtered beam spectrometer (RFBS)⁷ and the resonance filtered detector spectrometer (RFDS) belong to this category. Each subtype has merits and demerits. The RDS can give better statistics than the other two, which is the major advantage of the instrument. On the other hand, RFBS and RFDS are very simple to build, and easily adaptable to a multi-counter layout. The latter advantage is a disadvantage of a RDS, because γ rays from different foils at different scattering angles interact with each other. Present performance of these

instruments is already good enough for experiments measuring the spectra of scattered neutrons at large Q and large ϵ at large scattering angles.

For example, to perform an inelastic scattering measurement from superfluid ^4He using an RDS, one can expect to detect a well resolved condensate peak with a ratio $\Gamma_0/\Gamma_1 \sim 0.1$. Here Γ_0 is the width of the scattered neutron spectrum from the Bose condensate He and Γ_1 is from the normal liquid He. Here it is assumed a ^{238}U foil at 30 K is the resonance foil, at $\theta = 120 \sim 170^\circ$, with $L_0 \sim 10$ m and $L_f = 1 \sim 2$ m. In this case $Q = 120 \sim 150 \text{ \AA}^{-1}$, the recoil energy $E_R = 7 \sim 11$ eV and E_0 becomes about $14 \sim 18$ eV.

Present performance of this type instrument, however, is not sufficient to perform experiments in which Q is small but E_0 large. The signal to background ratio is not yet good enough. Significant improvements are necessary, and such efforts are in progress at various laboratories. In order to improve the energy resolution, some different methods have been proposed. Resolutions of $\Delta E \sim 35$ meV seem to be obtainable by taking a difference spectra obtained by different resonance detectors (different temperature, different thickness, etc.).

C. MAX Type Spectrometer

In the MAX type spectrometer, a crystal analyzer is placed after the sample to distinguish scattered neutrons of specific energy. TOF is used to measure the E_0 's of the neutrons that scatter from the sample down to these specific energies. In the lower $\hbar\omega$ range, a MAX type spectrometer becomes important because the measurement along a desired symmetry axis becomes important. Furthermore, one can perform polarization analysis by simply replacing the analyzer crystal with a Heusler alloy, combined with a white epithermal neutron polarizer. The flexibility of a MAX spectrometer, of course, is not as good as a triple axis spectrometer at a reactor, but the measuring efficiency for low energy spin waves ($\hbar\omega \sim 10$ meV) was proved to be almost the same as that with a triple axis spectrometer at a reactor, if the peak flux during each pulse is the same as the flux at a reactor, and if the measurement is over the entire Brillouin zone. This type instrument may be quite effective at energies of 100 meV to 300 meV.

D. TOF + Fixed-Low-Energy-Passer Spectrometer

Instruments in this category are inverted geometry type and utilize a low energy band passer to detect the scattered neutrons at fixed energy. Crystal

analyzer spectrometers CAT at KENS (time-focusing type), CAS at IPNS (E_f focusing type) and the FDS (Be-BeO filter difference spectrometer) at WNR belong to this category. In these spectrometers, Q is always nearly equal to k_0 because E_f is small. However, the instrument seems to be still very useful in some classes of measurements, especially because of its high efficiency and high energy resolution. Experiments for which this instrument works well are incoherent scattering such as hydrogen vibrational modes in metals, parametric studies of material (composition, temperature, etc.) and very dilute systems of light atoms in heavy hosts.

For example, the CAT type spectrometry can realize higher resolution $\Delta\hbar\omega/\hbar\omega < 1\%$ easily over the energy transfer range $100 < \hbar\omega < 1000$ meV with $L_0 \sim 10$ m, and a Be-Be' filter difference spectrometer (different Be temperatures) can realize much better resolution at $\hbar\omega < 150$ meV, because in this $\hbar\omega$ region, the main contribution to $\Delta\hbar\omega/\hbar\omega$ is ΔE_f .

E. Other Possible Spectrometer Types

The types of spectrometers discussed in the preceeding paragraphs have been tried and shown to be effective for measuring high energy excitations. A number of types of spectrometers have been proposed but not yet tried and these need to be tested. These spectrometers in diminishing order of promise and understanding are:

Constant Q Spectrometer (in testing stage at WNR and Harwell)

Polarizing filters (under development at SNS)⁸

Crystal monochrometers⁹

Rotating crystal chopper (used at reactors)

Resonance Bragg monochrometer

Resonance guide

Epithermal spin echo analyzer

Spin precession analyzer

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Figure 1. The Q, ω space accessible to the present neutron sources (below the $E_0 = 100$ meV line) and to the bench-mark pulsed spallation source (below the $E_0 = 10$ eV line).

Fig. 2. Crystal-field transition between the Γ_7 and Γ_8 states in BaPrO_3 at 260 meV as measured on HRMECS. The incident energy was 800 meV.

Fig. 3. The time-integrated neutron flux at the source for the two benchmark neutron sources, a 10^{16} peak rate pulsed spallation source and a 5×10^{15} flux reactor with hot source.

Fig. 4. Neutron current on target for similar capability spectrometers at the two benchmark neutron sources. The energy resolution selected was 1%.

FUNDAMENTAL NEUTRON PHYSICS

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I. Introduction

Over the last decade, a wide variety of intellectually tantalizing and fundamentally important physics experiments have been carried out on both the particle and wave properties of the neutron. In addition, reactor-produced intense γ -ray sources have been used in experiments yielding precise information on the neutron mass, γ -ray wavelength scale, and experiments are underway designed to yield certain combinations of the fundamental constants of nature.

This experience has led to various proposals for experiments requiring vastly expanded sensitivity, precision and sophistication. However, many of these ideas require an experimental environment which is somewhat less "hostile" in terms of, say, temperature stability and vibration isolation, than is commonly found around reactors. This is especially important, for example, in efforts to develop very large scale neutron interferometers in which the individual components of the device would be separately and dynamically positioned to a mÅ and oriented to a millisecond of arc. In order to obtain this "quiet, friendly" environment, a vibration isolation pier structure at the experimental station should be designed into the reactor building at the outset. For intense γ -ray source production and utilization, special remote manipulation and transport devices are necessary, which can only be built in an optimized fashion when directly coupled to the special needs for irradiation source volumes and beam hole geometries provided in the original facility design.

Most of the experiments envisioned require more-or-less permanent installations and the utilization of a neutron or γ -ray beam over extended periods of time, typically for several months, and in special cases for a year or more, for the full completion of a single measurement. Although the number of users of a given beam station may be smaller than for a typical neutron scattering spectrometer, the utilization is intense and basically full-time on the part of the participating scientists.

This report contains three main sections:

1. Neutron particle properties.
2. Neutron wave properties.
3. Fundamental physics utilizing reactor-produced γ -rays.

We have not explicitly dealt with conventional nuclear physics applications of reactors or spallation sources, neutrino physics or positron physics, due in part to limitations in time. These areas of science are equally important to those on which we have focussed our attention, and input from experts in these fields should be separately solicited in the planning and design stages of any new neutron source.

In this report we have first attempted to concisely summarize the current status of the science in the above three areas, and then to select those experiments on which we believe a new advanced high flux neutron source will make a qualitative impact.

II. Neutron Particle Properties

The past decade has seen significant activity and progress in the study of the particle properties of the neutron itself. Excellent reviews of this work exist in the form of workshop proceedings from the ILL (Van Egidy, 1978; Gönnawein, 1984). While virtually all of the experiments thus far carried out at reactors would benefit from increased flux, there appear to be some experiments which are likely to retain substantial interest through the next decade and would particularly benefit from increased neutron fluxes.

A. Neutron Decay

Neutron β -decay provides the prototype system in which to study semi-leptonic weak interactions. The fundamental parameter to be determined is the neutron lifetime τ_n . The uncertainty in this quantity currently limits our knowledge of the weak coupling constants and is of central importance in nuclear physics, astrophysics, and cosmology. In recent determinations, the current experimental limits are at the few percent level due primarily to systematic effects. Improvements of perhaps one order of magnitude are likely to be carried out on existing neutron sources. Beyond this point, statistical limits will become significant and further improvement will likely require improved source fluxes.

Related to τ_n are the various correlation coefficients in the decay of polarized (and unpolarized) neutrons. Sufficient accuracy in the determination of these quantities can (via the vector-axial (V-A) theory of weak interactions) provide an independent determination of τ_n . More significantly perhaps, a detailed study of the energy dependence of these correlation

light on higher order corrections to the V-A theory. Such detailed studies will likely be possible only at very intense neutron sources. Efforts to detect T-odd correlations in neutron β -decay have also been made and are continuing.

To summarize, we now anticipate significant progress in this area on existing sources in the near future. Much of this progress will result from improvements in experimental techniques. Ultimately, progress in these efforts will be limited by available fluxes. This limitation will likely occur at a level (perhaps 0.1%) beyond which interesting fundamental questions remain. Access to improved sources will then be of critical importance.

B. Neutron-Nucleon Parity Violations

Parity non-conserving (PNC) effects in nuclear phenomena are generally thought to arise from the electro-weak interaction between the quarks within nucleons. While there is no particular expectation that the Glashow-Weinberg-Salam theory will not apply to quark-quark interactions within nucleons, the standard model has not been extensively tested in this area.

Existing parity violation data in nuclear physics comes primarily from effects in relatively complex nuclei. While considerable theoretical progress has been made in the interpretation of this data, direct tests in simple systems are highly desirable. There are two particular experiments which have been shown to be feasible, but they are currently limited by flux considerations from reaching sensitivities which could test fundamental theories.

The first of these is the asymmetry in the emission of γ -rays following the capture of polarized neutrons by protons. Fundamental theories predict an asymmetry of $\sim 6 \times 10^{-8}$. The current error of 5×10^{-7} is statistically limited, and it is unlikely that a detailed test of the theory can be made with existing sources.

The second phenomena is the parity non-conserving spin rotation of the neutron. An experiment to measure this effect in the n-p system would provide information complimentary to the gamma asymmetry experiment, and the two results would determine the phenomenological parameters describing n-p PNC effects. The expected rotation of $\sim 10^{-8}$ rad/cm would be detectable with an improved neutron source.

In addition to the n-p systems, investigation of PNC effects in simple systems, n-d, n-³He, and n-⁴He could provide important, interpretable information.

C. Neutron-Neutron Interaction

The fact that the neutron interacts strongly with other hadrons is so evident that one might suppose that there is little of a fundamental nature to be investigated with low energy neutrons. However, one interaction which has not yet been directly investigated is the neutron-neutron strong interaction. Indirect experiments involving $\pi+d \rightarrow n+n+\gamma$ and $n+d \rightarrow n+n+p$ can shed some light on this question, but the interpretation of such experiments is complicated by additional strong interactions. A direct determination of the n-n scattering length would be an important result and would have particular implications concerning the charge independence of nuclear forces.

A limiting source of background in such an experiment arises from spurious events between the neutron beam and background gas in the interaction volume. A necessary requirement for the feasibility of such an experiment is that one can obtain a neutron density which is comparable to, or less than, the background gas density. This condition is marginally satisfied at the HFR at ILL. Since n-n reactions will proceed at a rate proportional to the flux squared, there is a clear advantage to an advanced, more intense, neutron source.

This interesting experiment would be difficult even at a new generation neutron source. However, with such a source it becomes possible. We estimate that with a reactor source of $\phi = 10^{16}$ n/cm²/sec, a neutron-neutron scattering rate of 1/sec within a 1m x 10cm x 10cm interaction volume along an external beam is achievable.

A very intense pulsed neutron source, at a peak flux of say 10^{17} n/cm²/sec, may provide a different, unique opportunity for measuring the n-n scattering cross section. One could envision utilizing cold or even ultra-cold neutrons produced in one pulse as the "target" and the next thermal neutron pulse as the "incident beam." Phase sensitive detection of the scattered neutrons would then provide an inherently, very low background situation. We have not attempted to analyze this concept quantitatively.

D. Neutron EDM, and n - $\bar{n}$ Oscillations

Of the number of particle physics questions to be addressed at an advanced neutron source, perhaps the two foremost are the searches for the electric dipole moment (EDM) and neutron-antineutron oscillations (n - $\bar{n}$). Both experiments would provide information leading to extensions of the highly successful, but obviously incomplete, gauge theories that describe elementary particle properties and interactions. Just as tests to confirm this "standard model" have occupied experimental particle physics for the last fifteen years, the search for clues to extend this picture will likely occupy particle physicists for the next fifteen years. Neither of these searches has yielded a positive result thus far, but a higher flux neutron source can be expected to extend the sensitivity for each by perhaps an order of magnitude.

The existence of the neutron EDM would be an example of CP or T-symmetry violation. Only the K^0 - $\bar{K}^0$ system has revealed such a violation, and its origin in nature has remained a mystery since its discovery. CP violation follows naturally from a number of particle physics theories suggesting its source to be the electromagnetic, or weak interaction, heavy gauge particles, imaginary mixing phases, or totally new super-weak interactions. A profound suggestion links CP-violation to the baryon-antibaryon asymmetry of the universe. There are a number of theoretical predictions just below presently available experimental limits, which would be within reach at a higher flux source.

The n - $\bar{n}$ experiment searches for a $\Delta B=2$ interaction which could arise in a similar manner as the $\Delta B=1$ reaction searched for in proton decay experiments. In fact, an n - $\bar{n}$ effect would be complimentary to proton decay giving evidence for "right-handed" interactions in an otherwise dominantly "left-handed" universe. At present there is a controversy whether bound neutrons could oscillate, such that the current round of proton decay experiments might already have set limits on its existence. It appears, however, that a search using free neutrons would be more sensitive as the effect can be disrupted by a small perturbing magnetic field, let alone the forces involved in a nucleus.

EDM: The most sensitive method known to measure the EDM uses a split coil nuclear magnetic resonance device in which a neutron bottle storing ultra-cold neutrons (UCN) is inserted to increase the measurement time (see the review by Golub and Pendlebury (1979), and the article by Ramsey (1981)). Current limits are $4\text{--}5 \times 10^{-25}$ e-cm obtained in reactor experiments

at Leningrad and Grenoble. Improvements anticipated at existing neutron sources could lower this limit to 10^{-26} e-cm within a few years. For additional improvements below this value, not only will a more intense source of neutrons be necessary, but also technological improvements must be sought to reduce systematic effects, such as the use of a superconducting magnetic shield to reduce magnetic field inhomogeneities.

The EDM experiments will require a cold moderator and perhaps a direct UCN converter within the neutron source with an efficient guide tube to transport the neutrons to the bottle. A magnetically quiet environment near the EDM apparatus would also be required.

During the last five years, two new ideas have been demonstrated to work in producing UCNs:

- a. Down-scattering of cold neutrons in liquid helium-4 to ultra-cold velocities at the ILL.
- b. Doppler-shifting of cold neutrons to ultra-cold-velocities by Bragg scattering from moving mica crystals at Argonne.

Both of these techniques appear to hold considerable promise in the next generation of EDM experiments. The second technique can take advantage of the extremely high peak fluxes projected for the pulsed spallation sources, since a UCN bottle can (in principle) be filled to the corresponding peak neutron density (as opposed to the time averaged density).

$n-\bar{n}$: An experiment at Grenoble has placed an upper limit on the oscillation time at 10^6 seconds. A proposed improved experiment could yield a two order of magnitude improvement on the current result. There are predictions of $n-\bar{n}$ oscillations which fall within the sensitivity of this experiment, but the gauge theories say that an experiment with 10^9 second sensitivity would be more definitive. This should be within reach of an experiment at an advanced neutron source relying on statistics alone.

The experiment uses a long flight tube shielded from the earth's magnetic field. Any converted antineutrons will be absorbed on a target at the end of the tube and the 2 GeV of released energy is detected in a calorimeter detector surrounding the target. To reduce cosmic ray backgrounds which ultimately limit the sensitivity, a tracking detector to isolate events to those originating in the target is necessary. Due to the size and complicated design, the detector will be expensive.

As the sensitivity varies as flight-time squared, colder neutrons will yield lower limits. Thus, cold or even an ultra-cold neutron source is required. The size of the detector and its required shielding will add other, special constraints on the experimental area.

III. Neutron Wave Properties, Optics, Crystal Interferometry

A variety of phenomena analogous to those of classical optics are known to be exhibited by slow neutrons. Substantial interest in this field has been generated in the last 5 to 10 years from the point of view of fundamental physics. This has been due to largely to the successful utilization of the perfect crystal interferometer and to the increased availability of more intense beams of cold neutrons (see the review by Klein and Werner, 1983). These perfect crystal devices have allowed the observation of quantum-mechanical interference phenomena on a macroscopic scale. They are sensitive to quite small potentials; the current level is about 10^{-12} eV.

A. Neutron Michelson-Morley Experiment

The interferometers currently in use are monolithic, achromatic devices having an enclosed area (Fig. 1a) and are therefore sensitive to rotations (Sagnac effects) but not independently to translations. Since the coordinate transformation properties for neutrons and photons are different, carrying out a Michelson-Morley experiment with neutrons (preferably simultaneously with X-ray photons) is of interest. A schematic diagram of a perfect crystal "Michelson-type" interferometer is shown in Fig. 1b. This is a chromatic, dispersive-type of interferometer which reduces the corresponding counting rates by a factor of about 1000. Thus, neutron sources of 10^{15} , or perhaps 10^{16} n/cm²/sec will be essential in carrying out a serious experiment of this type in which the diurnal and annual relative phase shift of X-ray photons and thermal neutrons are measured.

If the back-reflecting corner crystals are separated from the incident beam splitting crystal (and then individually positioned and oriented) creating very long beam paths, say of order 1m, the overall sensitivity of this interferometer could be substantially greater than the current monolithic Sagnac interferometers. The sensitivity is then sufficient to observe the phase shift due to adjacent massive (dense) objects (such as a cylinder of

Uranium metal)-a neutron-Cavendish experiment. These long beam paths would also be of potential value in parity-violating rotation experiments during the passage of neutrons through matter, and perhaps also in the measurement of the neutron's electric polarizability.

B. Large Coherence Volume Interferometers: Hanbury-Brown-Twiss Experiment

Up until now, all perfect crystal interferometers use crystal collimation only in one direction, which is of the order of the ratio of the lattice constant deviated by the thickness of the crystal (a/t). With this quantity and the uncertainty relation, the related transverse coherence length is given by the width of the Borrmann fan $\Delta_H = 2t \tan \theta_B$ which is of the order of several mm. The longitudinal coherence length is given by the wavelength spread as $\Delta_L = \lambda^2 / \Delta \lambda$ and reaches for high resolution systems about 2000 Å. Due to the lack of an effective vertical collimation (~ 20 min of arc) the vertical coherence length is rather small ($\Delta_V = 20$ Å). A crossed perfect precrystal can be used to increase this to $\Delta_V = \Delta_H$. In this case a coherence volume of about 10^{-6} cm^3 can be achieved which is at the limit where one could conceive of using a very thin detector for a Hanbury-Brown-Twiss experiment; that is, a coincidence measurement between beam paths I and II. For an estimate of the reduction of the intensity compared to presently operating interferometers, one has to take into account the following factors:

- high resolution (factor 20),
- vertical collimation (factor 600),
- efficiency of a 2000 Å thick detector (factor 100),

and the fact that the expected Hanbury-Brown-Twiss dip of the coincidence counting rate has a width of $\Delta t = \Delta_V / v$, which is on the order of 0.1 nanosecond. From these numbers, one expects for a $10 \times 10 \text{ cm}^2$ beam, a coincidence counting rate (at a $10^{16} \text{ n cm}^{-2} \text{ s}^{-1}$ source) of about 1 neutron per week. Perhaps additional bunching methods described later can increase this number by a factor of 10. Therefore, it seems worthwhile to look at the feasibility of such a new type of matter wave optics experiment in greater detail. The larger coherence volume will be, in any case, useful for new kinds of coherence measurements like quantum chopping, phase echo systems, etc.

C. Neutron Electric Polarizability

The internal quark structure of the neutron predicts a finite electrical polarizability of the neutron which is of the order of $\alpha = 10^{-3} \text{ fm}^3$. This gives rise to an interaction term within an electric field of αE^2 . This gives within the Coulomb field of heavy nuclei a contribution to the scattering length of $\sim 0.04 \text{ fm}$, which is certainly measureable, but it cannot be separated from the nuclear interaction. With present monolithic interferometers and achievable laboratory fields, the effect on the neutron phase is too small to be detected. With the advent of large scale interferometers with interaction lengths of the order of meters, a direct measurement becomes feasible. Even then the phase shift would be only 10^{-5} rad/m for a field of 10^5 V/m . However, in a very "quiet" reactor environment, one could consider multiple-pass interferometers in which the beams in each of the two legs of the interferometer are reflected back and forth between perfect Si crystals n -times to enhance the interaction time.

D Active Neutron Optical Devices

Neutron optical components have been developed which shift neutrons from a certain phase space element into another to achieve novel bunching or focussing effects. Devices of this type fall into two categories: the first act directly on the neutron intensity spectrum. Examples are totally reflecting turbine blades and Bragg-reflecting crystals mounted on rotating wheels. They have been shown to be useful in producing ultra-cold neutrons by Doppler-down-shifting cold neutron beams. They will be used frequently at new facilities, provided that their optimal layout is considered from the initial source planning stages. The second category of these devices act "phase-non-destructively" on the neutron wave function. Their action can be described completely by a Hamiltonian function.

The neutron magnetic resonance system represents such a device (Fig. 2a). At resonance, the Zeeman energy ($\Delta E = \pm 2\mu_B$) is exchanged between the neutron and the oscillating magnetic field providing an energy separation behind the system of $4\mu_B$ ($\sim 0.5 \mu\text{eV}$ for a field $B = 20 \text{ kG}$). This effect has been verified experimentally in 1980 using a backscattering spectrometer at Jülich and in 1983 in Japan for ultra-cold neutrons (see the paper by Rauch, 1983). By means of a multistage system, it can be used for unique beam tailoring (Fig. 2b). The system is designed to pump neutrons into a specific small energy interval. The neutron magnetic energy shifts match well the

dynamical reflection widths of perfect crystals, and therefore their use within such a multistage scheme provides novel opportunities for beam bunching and focussing. The use of superconducting magnets can substantially increase the efficiency by increasing the energy shift at each stage. The use of a gradient field can provide a high flipping probability, even for a very divergent beam (~ 20 deg), which would be essential in the ultra-cold neutron regime.

New aspects of this multistage, electromagnetic system appear if the incident beam has a pulsed structure. In this case, the system can be operated with a time-dependent structure to achieve an effective bunching of the neutrons into specific energy or time intervals. The technology for fast-flipper-chopper devices has been demonstrated in part, and has only to be adopted for this purpose. In addition to these resonance devices, time-dependent magnetic field components are currently being discussed. They are similar in certain respects to magnetic-kicker systems for charged particle accelerators.

The main application of these electromagnetic beam tailoring systems will be in high resolution neutron scattering work and is the field of ultra-cold neutron research.

IV. Fundamental Physics Utilizing Reactor-produced γ -ray Sources

Gamma ray spectra, especially as can be obtained using sources internal to high flux reactors, are significant in the general area of nuclear structure studies and, in special cases, for the determination of fundamental physical constants and the neutron's mass. While nuclear structure studies are well supported by modestly high resolution instrumentation (provided a consistent energy scale is available), the fundamental measurements are more demanding in terms of resolution. They require, as well, a robust linkage to optical standards, e.g., the hydrogen Rydberg or the electromagnetic scale's defined "standard." (See the references, Deslattes, et. al (1980,1983)).

General spectroscopic measurements have been (and will continue to be) carried forward by means of large radius (~ 25 meter) focussing spectrometers. These are basically comparison instruments which relate spectral lines under study to secondary standards otherwise established either via fundamental constants such as electron-positron annihilation or using mass

spectroscopic normalization or, most recently, by direct optically based measurement.

The nuclear spectroscopy work mentioned above is exemplified by the long standing program at the ILL which uses a dual 25 meter instrument, GAMS 2,3, and a small 10 meter system, GAMS 1. These spectrometers are designed so as to be able to view small sources in a through-tube tangent to the reactor core. A manipulator system makes it possible to introduce and remove sources during reactor operations and position them in a region of high thermal flux. Performance obtained using GAMS 2,3 is limited by the source size and its motion, as well as characteristics of the bent crystals. These tend to restrict both available resolution and one's ability to work at energies near 2 MeV and above.

Within the past decade it has become possible to generate energy (wavelength) values for secondary gamma ray standards using a new procedure which is both rather direct and capable of high accuracy. This procedure (which has been described elsewhere) begins by optical interferometric measurement of the lattice period in a highly perfect silicon single crystal. The calibration thus established is then transferred to other crystals of Si and Ge using an effectively non-dispersive procedure. These are subsequently used in interferometrically controlled transmission double crystal instruments to generate accurate wavelength values via the Bragg-Laue condition, $\lambda = 2d\sin\theta$. These flat crystal instruments are needed in order for measurements to proceed using the Bragg-Laue relation but suffer from extremely low efficiency. They thus require intense sources ($\sim 10^3\text{Ci}$ or more, if possible). Early work was confined to sources having sufficiently long half-lives that they could be removed from the reactor in which they were activated and brought to an external area for measurement. This restricted work to $E_\gamma < 1$ MeV. Most recently such instrumentation has been established and operated at the ILL permitting the range of optically based secondary standards to be extended above 2 MeV for the first time. This instrumentation, GAMS-4, is located on the reactor floor in a specially constructed blockhouse.

Initial results are encouraging in several ways. Firstly, certain crystals, especially one pair of Ge (400) specimens, have continued to behave according to dynamical diffraction theory even to the highest energies and highest orders studied. Diffraction patterns obtained (see Fig. 3) indicate

that a spectroscopic resolving power $E/\Delta E \sim 3 \times 10^5$ is available up to 2 MeV and likely beyond. This constant resolving power is (in a flat-crystal instrument) necessarily accompanied by a solid angle of acceptance which decreases as E^{-1} . This behavior, when combined with the source size (volume) restrictions in the ILL facility, means that the full potential of this type of instrumentation in the region $E_\gamma > 2$ MeV will not be realized at the ILL.

This last point is particularly unfortunate from the point of view of fundamental physics. Firstly, the neutron's mass follows from n-p capture at 2.2 MeV. With some planned source carrier modifications, this can be treated adequately for most purposes except for the following consideration: Measurement of the q-cascade in $^{14}\text{N} + n$ combined with the n-p energy gives a binding energy difference between ^{15}N and ^2H for which there is a corresponding mass doublet between $^{15}\text{NH}_3$ and $^{14}\text{NH}_2\text{D}$ that can be measured quite well. If one writes $\lambda^* = hc/E$ then $\Delta M^* \lambda^* = N_A h/c$, a new combination of physical constants becomes accessible. But this is only possible if high accuracy measurements can be extended to the region 4-5 MeV. Progress toward this goal can be made over the next several years, but the full potential can be reached only if a high flux reactor can be equipped with a source manipulator appropriate to the special needs of flat crystal instrumentation.

Overall, we are impressed by the potential gains which become possible if a state-of-the-art reactor were to be equipped for q-ray spectroscopy with designs for source manipulation, and spectrometers based on experience obtained in the past decade. In the case of curved crystal instrumentation, increasing the minimum radius of curvature to 50 meters would immediately solve many problems as would a positive positioning system for the source and/or active position monitoring. Such instruments require anchoring to the reactor vessel and/or an optical keel. Provision for large volume sources (especially depth along the line of sight) would make the full potential of flat crystal instrumentation accessible. This type of spectrometer must be decoupled from environmental noise and vibration. They must be thermally stabilized, but they do not require critical pointing with respect to source position. A new class of crystal-diffraction spectrometer is under development with intermediate characteristics between those of double-flat-crystal spectrometer and single-bent-crystal instruments. These will be double-curved-crystal devices with average radii near 100 meters. Their pointing and environmental requirements will likewise be of intermediate

character. Such devices should recover most of the efficiency loss entailed in going from curved crystal instruments to flat crystal instruments but are much less dependent on source position and pointing errors than are the curved crystal devices.

V. Concluding Remarks

In the introduction, it was stated that we have focussed our attention here on only three areas of fundamental physics at reactors (or spallation neutron sources), namely: neutron particle properties, neutron wave properties, and a selected class of γ -ray experiments. We have, by no means, exhausted the interesting ideas for experiments in these three areas that could potentially benefit from a new, very high flux neutron source. For example, we have not discussed the neutron interferometry versions of Wheeler's delayed-choice, two-slit experiments, or the higher-order gravitational phase shift experiments suggested in recent years by J. Anandan and L. Stodolsky. However, the biggest omissions are the many experiments in nuclear structure, positron and neutrino physics that such a potent new neutron source would impact.

Nevertheless, we trust that the basic message we have conveyed in this short, three-day study is inescapable:

1. There is a wide spectrum of fundamental questions in physics to be addressed with new advanced neutron facilities and instrumentation.
2. Special facility design considerations are extremely important to implement in the initial source construction phases in order to optimize the probability for success in any of these rather complex and delicate experiments.
3. Most of the experiments in this area of physics would be totally compatible with, and probably beneficial to, a large (and perhaps dominant) neutron scattering program at a new high intensity neutron source.
4. Success in certain experiments discussed here could substantially effect our fundamental view of nature, and thereby justify, ex post facto, a

large fraction of the cost of such an expensive source.

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Fig. 1. Schematic diagram of neutron interferometers. (a) Achromatic interferometer as presently in use, (b) Chromatic interferometer.

Fig. 2. NMR systems as applied to neutron beam interactions. (Single stage, (b) Multistage for beam tailoring.

Fig. 3. Intensity versus crystal angle from a germanium flat crystal for a 199 keV γ -ray. Experiments conducted at the Institut Laue-Langevin.

VII. PARTICIPANTS AND ACKNOWLEDGMENTS

A total of 61 scientists participated in the Shelter Island Workshop. Of these 29 were affiliated with the five major U.S. neutron facilities, 10 came from U.S. or Canadian Universities and institutions, 20 came from Europe, and 2 from Japan. An alphabetical list is attached as Appendix 3 to this report and the scientists in each group are listed at the top of each section.

The editors wish to acknowledge the fine job done by all the individual group chairmen in leading the discussions and getting their material in within a reasonable time. Kent Crawford of Argonne assisted in the early planning of the Workshop and the preparation of Appendix I, but illness prevented him from attending the meeting itself. On behalf of all of us we wish him a speedy recovery. Thanks are also due to administrative staff at Argonne, Jeannie Glover, and at Brookhaven, Nancy White and Patricia Tuttle, who helped to make the Workshop a success. Special thanks from all the participants to the proprietor and staff of the Pridwin Hotel on Shelter Island who provided a series of elegant meals in a fine fall setting. Support for the Workshop was provided by the Department of Energy, the Department of Commerce, and the University of Chicago and we express our thanks to all of them.

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APPENDICES

The following two documents were sent out to all participants of the Workshop before the meeting, and are reproduced here for completeness.

Background Information on Advanced Spallation Source

R. K. Crawford (ANL), G. Russell (LANL), and J. Carpenter (ANL)

The time-averaged neutron flux per unit energy on a sample, monochromator, or other device at a distance L from the source is

$$(1) \quad \Phi(E) = I(E)/L^2$$

where $I(E)$ is the neutron beam current, expressed in neutrons/steradian-sec, from the entire viewed face of the moderator ("viewed source"). For small or "fast" moderators tailored to produce relatively "short" pulses at pulsed sources, the beam current is approximately

$$(2) \quad I(E) = I_{th} \left[\frac{E}{E_t^2} e^{-E/E_t} \right] + I_{epi} \left[\frac{\Delta(E)}{E} \right] \left[\frac{E}{E_{ref}} \right]^\alpha$$

Here $\Delta(E)$ is a "joining" function which goes smoothly from 0 for E below about $5 E_t$ to 1 for E above about $5 E_t$, E_{ref} is a reference energy (typically taken to be 1 eV) and α is a small number ($0 < \alpha \lesssim .1$ for a typical moderator). The absolute magnitudes of I_{th} and I_{epi} are proportional to the accelerator proton current and roughly to the proton energy and to the mass number of the target material (with extra credit for fission!). These also depend upon moderator and reflector material and geometry, and on the target-moderator coupling. The characteristic energy E_t depends strongly on the physical temperature and material of the moderator, and for "good" moderators will be only a small amount above kT . (Sometimes a "poor" moderator may be the best choice for particular applications, however.) For some of the possible choices of moderators, both large and small, the "thermal" part of the spectrum is not Maxwellian, so Eq. 2 should be regarded as only a rough approximation.

It is also possible to use large or "slow", tightly coupled moderators at pulsed sources. In this case, the neutron pulses produced are much longer in duration (hundreds of microseconds) than are the pulses from the small, "fast" moderators (~ 1 -100 microseconds), but the time-averaged beam intensity produced is also much greater. In this "highly-moderated" case, the epithermal part of the spectrum is very much smaller, with much greater intensity developed in the

Maxwellian thermal part of the spectrum as in reactors, so only the first term in Eq. 2 is significant.

Two often used quantities for characterizing a pulsed source are the time-averaged and peak 4π effective fluxes. From the beam current given in Eq. 2 one can calculate an "effective 4π equivalent time-averaged total Maxwellian source flux" $\bar{\phi}_{\text{eff}}$.

$$(2) \quad \bar{\phi}_{\text{eff}} = \frac{4\pi}{A_{\text{moderator}}} \int_0^{\infty} I_{\text{Maxwellian}}(E) dE$$

$$= \frac{4\pi}{A_{\text{moderator}}} I_{\text{th}}$$

This is the source flux which a reactor would need in order to produce the same time-averaged neutron beam current, and is an indication of how effective the pulsed source would be for an instrument making no use of the pulsed source time structure. Similarly, one can define an "effective 4π equivalent peak total Maxwellian source flux" ϕ_{eff}

$$(3) \quad \phi_{\text{eff}} = \frac{\bar{\phi}_{\text{eff}}}{f\tau} = \frac{4\pi}{A_{\text{moderator}}} \frac{I_{\text{th}}}{f\tau}$$

where f is the source frequency and τ is a typical neutron pulse width. This "peak flux" is a quantity which very roughly represents the relative effectiveness of a pulsed source for instruments making maximum use of the pulsed source time structure.

The use of such "effective flux" quantities is somewhat controversial, and the literature abounds with reported effective fluxes based on somewhat different definitions. However, there is no need to resort to this effective flux concept at all. All that is necessary for the calculation of instrument performance is the time-average neutron beam current $I(E)$, which is the fundamental quantity calculated or measured for pulsed sources in any case, the pulse width as a function of energy, viewed source area, and source repetition rate. These quantities are detailed for a "state-of-the-art" source in the following section.

"State-of-the-Art" Pulsed Source

Two different approaches have been proposed for generating the large proton currents necessary for the next generation of pulsed spallation neutron sources.

These are the linear accelerator (5 mA time-averaged proton current at 1.1 GeV) proposed as the basis for the German SNQ project, and the fixed-field alternating-gradient synchrotron (3.8 mA time averaged proton current at 1.5 GeV) studied at Argonne as the basis for the ASPUN project. Although the SNQ and ASPUN involve very different accelerator systems, these systems as proposed would result in almost identical time-averaged neutron production rates. Since after extensive review these systems seem feasible, it is reasonable to take this level of neutron production as "state-of-the-art".

On this basis, a "state-of-the-art" pulsed spallation neutron source is suggested for use in the calculations at this Workshop. For the present calculations, 5 different moderators are defined.

Small Decoupled, "Fast" Moderators

A: 300 K liquid H_2O

B: 100 K liquid CH_4

C: ~20 K liquid H_2

Large, Coupled, "Slow" Moderators

D: 300 K liquid D_2O

E: ~20 K liquid D_2

Moderators A, B, and C should each be assumed to have a viewed surface of $10 \times 10 \text{ cm}^2$. Moderator D is a large ($\geq 1 \text{ m}^3$) tank of D_2O , and moderator E is a smaller cryogenic moderator embedded in the D_2O of moderator D. Both moderators D and E would contain reentrant beam tubes and/or guides, similar to the case at a reactor. For the present calculations, these beam tubes should be assumed to have cross-sectional areas of about 100 cm^2 , but this can be adjusted (within limits) to match specific instrument requirements.

Figure 1 represents estimates of the energy dependent time-averaged beam current (based on 100 cm^2 viewed source) to be expected from each of these moderators at this source. Figure 2 contains the corresponding expected pulse widths. For reference (but not needed for calculational purposes), Table I shows the total time-averaged 4π equivalent Maxwellian fluxes expected for each of these moderators.

All necessary source parameters for instrument calculations can be obtained from Figures 1 and 2. It should be assumed that the source operates at 50 Hz so any instruments requiring a lower repetition rate will suffer a corresponding loss in time-averaged intensity. (A lower repetition rate can be achieved by the use of pulse-removing neutron choppers, or by multiplexing the proton beam between two target stations, one of which operates at the lower frequency.)

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Table I

"State-of-the-Art" Pulsed Source Moderators

Moderator	Size	Type	Temp.	$\bar{\phi}_{\text{eff}}$
A	10x10x5 cm ³	Poisoned, Decoupled, H ₂ O	300 K	1.3 x 10 ¹⁴ n/cm ² -sec
B	10x10x5	Poisoned, Decoupled, CH ₄	100	7.9 x 10 ¹³
C	10x10x5	Decoupled H ₂	20	8.0 x 10 ¹³ *
D	~ 100x100x100	Coupled D ₂ O	300	5.3 x 10 ¹⁴
E	~ 30x30x30	Coupled D ₂	20	1.8 x 10 ¹⁴ *

* Approximate estimate since spectrum is non-Maxwellian.

Fig. 1. Time-averaged energy-dependent neutron beam current $I(E)$ for the state-of-the-art pulsed spallation neutron source moderators. A "viewed source" of 100 cm^2 is assumed for all moderators.

- Moderator A: Time structure based on IPNS measurements (Carpenter and Ikeda, 1984) for CH_2 . Spectral shape based on SNS calculations (Taylor and Picton, 1984). Absolute intensity based on mock-up calculations (Russell, 1984)
- Moderator B: Time structure and spectral shape based on calculations for SNS moderators (Taylor and Picton, 1984). Absolute intensity based on mock-up calculations (Russell, 1984)
- Moderator C: Time structure based on ZING-P' measurements (Carpenter, et al., 1981). Spectral shape based on ZING comparison of coupled and decoupled H_2 (Carpenter, et al., 1981) and SNS calculations (Taylor and Picton, 1984) for coupled H_2 . Absolute intensity based on mock-up calculations (Russell, 1984)
- Moderator D: Time structure based on SNQ mock-up measurements of the time decay constant for low energy neutrons (Bauer, et al., 1981), and on the infinite medium value for neutrons above 1 eV. The curve connecting these two points is a guess. Spectral shape is assumed to be Maxwellian plus a $1/E$ epithermal term, with the ratio of these components based on ILL flux distribution data (Ageron, 1972). Absolute intensity is based on SNQ mock-up measurements (Bauer et al., 1981), approximately adjusted for use of Ta target.
- Moderator E: Time structure is assumed to be the same as for moderator D (no data available). Spectral shape and intensity relative to moderator D are based on data for the ILL cold source (Ageron, 1972)

Fig. 2. Pulse widths for the pulsed spallation neutron source moderators.

BACKGROUND INFORMATION ON NEUTRON FLUX
FOR AN ADVANCED HIGH-FLUX REACTOR

R. M. Moon, Oak Ridge National Laboratory

I. SOURCE FLUX

We assume a large D₂O reflector equipped with cold source(s), a hot source, and with a peak thermal flux in the reflector of $\phi_0 = 5 \times 10^{15} \text{ n cm}^{-2} \text{ sec}^{-1}$. In this section we estimate the flux distributions $\frac{d^2\phi}{d\Omega d\lambda}$ ($\text{n cm}^{-2} \text{ sec}^{-1} \text{ sterad}^{-1} \text{ \AA}^{-1}$) for thermal, cold, and hot beams.

A. Thermal Distribution

We assume an isotropic, Maxwell-Boltzmann distribution.

$$\frac{d^2\phi}{d\Omega d\lambda} = \frac{\phi_0}{4\pi} v_\lambda(T_0, \lambda) \quad (1)$$

$$v_\lambda(T_0, \lambda) = \frac{2}{\lambda} \left(\frac{E}{E_0} \right)^2 e^{-E/E_0} = 2 \lambda_0^4 \lambda^{-5} e^{-(\lambda_0/\lambda)^2} \quad (2)$$

$$\lambda_0[\text{\AA}] = 30.81/\sqrt{T_0[^\circ\text{K}]} \quad (3)$$

$$E_0[\text{meV}] = 0.08616 T_0[^\circ\text{K}] \quad (4)$$

Figure 1 shows a comparison between the observed distribution at the H12 thermal beam¹ at the ILL and a calculated distribution with $\phi_0 = 1.2 \times 10^{15} \text{ n cm}^{-2} \text{ sec}^{-1}$ and $T_0 = 310 \text{ K}$. These parameters give a fairly good agreement, particularly for $\lambda > 0.9 \text{ \AA}$. Accordingly, to describe the thermal spectrum at our assumed reactor, we use $\phi_0 = 5 \times 10^{15}$ and $T_0 = 310 \text{ K}$.

If it is desired to express the distribution in terms of an energy interval, the appropriate equations are

$$\frac{d^2\phi}{d\Omega dE} = \frac{\phi_0}{4\pi} v_E(T_0, E) , \quad (5)$$

$$v_E(T_0, E) = \frac{E}{E_0^2} e^{-E/E_0} . \quad (6)$$

Note that

$$\int_0^{\infty} v_{\lambda} d\lambda = \int_0^{\infty} v_E dE = 1 , \quad (7)$$

and that

$$v_{\lambda} = \frac{2E}{\lambda} v_E , \quad (8)$$

and that

$$\lambda[\text{\AA}] = \frac{9.044}{\sqrt{E[\text{meV}]} } . \quad (9)$$

B. Cold Distribution

The problem with cold source spectra is that they do not follow a Maxwell-Boltzmann distribution, as pointed out by Ageron et al.² Therefore, in estimating the cold neutron spectrum for our 5×10^{15} reactor we will rely on a scaled-up version of the ILL distribution. There is even a problem with this procedure in that the calculated ILL cold source spectrum^{3,4} is significantly higher than that derived from neutron guide measurements.¹ This difference is now completely understood by Ageron⁵ and is due to three effects: (1) the original calculation was based on unperturbed rather than the perturbed flux at the cold source position, (2) the calculation was based on gain factors measured with a larger D₂ volume than that in the HFR installation, and (3) the real guide transmission is not perfect. Recent calculations are in excellent agreement with the measured guide spectra.

According to Ageron³, the unperturbed thermal flux at the cold source position is $(4\pi)(3)10^{13} = 3.8 \times 10^{14} \text{ n cm}^{-2} \text{ sec}^{-1}$. We hope to install a cold source in a region with unperturbed thermal flux of $2-3 \times 10^{15} \text{ n cm}^{-2} \text{ sec}^{-1}$, so it is reasonable to use a scale factor of six applied to the ILL spectrum in making this estimate. The upper curve marked "cold A" in Fig. 2

is based on the original ILL calculation^{3,4} and the lower "cold B" curve is based on the measured guide spectra, assuming perfect guide transmission. If our purpose is a comparison with the pulsed source described by Crawford et al. in these proceedings, then the "cold A" curve should be used because they have also used this curve in generating the cold source spectrum for the case of a liquid D₂ source embedded in a large D₂O moderator. If our purpose is to compare with existing ILL instruments, then probably the "cold B" curve should be used. By changing the shape of the ILL cold source, Ageron estimates that the cold flux can be increased by a factor of 1.8. This "reentrant factor" has not been included in the curves of Fig. 2.

C. Hot Distribution

We base our hot source distribution of Fig. 2 on the H3 curve of Ref. 1 using a scale factor of four to go from a 1.2×10^{15} reactor to a 5×10^{15} reactor.

D. Summary

The hot, thermal, and cold distributions are given in Fig. 2. The hot distribution is based on the H3 curve of Ref. 1 with a scale factor of four. The thermal distribution is based on Eqs. (1), (2), and (3) with $T_0 = 310$ K and $\phi_0 = 5 \times 10^{15}$ n/cm² sec. The cold distributions are based on the ILL cold source with a scale factor of six. For comparison with the "state-of-the-art" pulsed source with a large D₂O moderator and a D₂ cold source, the curve labeled "cold A" should be used.

II. BEAM TUBES, GUIDES, AND MONOCHROMATOR SYSTEMS

In this section we summarize equations needed to calculate neutron intensities delivered by beam tubes and guides and present some expected intensities for the assumed 5×10^{15} reactor. In addition, the equations needed to calculate monochromatic intensities delivered to the sample by a crystal monochromator are presented.

A. Beam Tubes and Guides

$$I(\lambda) = \phi''(\lambda) \delta_H(\lambda) \delta_V(\lambda) \quad [n \text{ cm}^{-2} \text{ sec}^{-1} \text{ \AA}^{-1}] \quad (10)$$

where $\phi'' = d^2\phi/d\Omega d\lambda$ and δ_H and δ_V are the horizontal and vertical angular divergences of the tube or guide.

Beam Tube

For a beam tube of width w_S and height h_S at the source and of length L_0 , we have

$$\delta_H = w_S/L_0 \quad (11)$$

$$\delta_V = h_S/L_0. \quad (12)$$

Typical values might be $w_S = 10 \text{ cm}$, $h_S = 15 \text{ cm}$, and $L_0 = 700 \text{ cm}$.

Straight Guide (Long)

$$\delta_H = \delta_V = 2\theta_C = 2 K \lambda. \quad (13)$$

The constant K is listed below for various guide coating materials.

	<u>$K \text{ (radians } \text{\AA}^{-1})$</u>
Ni	$1.73 \cdot 10^{-3}$
^{58}Ni	$2.05 \cdot 10^{-3}$
Supermirror	$3.81 \cdot 10^{-3}$

Horizontally Bent Guide

The result of Maier-Leibnitz and Springer⁶ can be written as follows,

$$\delta_V = 2 K \lambda \quad (14)$$

$$\delta_H = \left(\frac{\bar{\omega}}{\omega_S} \right) \delta_V \quad (15)$$

$$\left(\frac{\bar{\omega}}{\omega_S}\right) = \begin{cases} \frac{2}{3} \left(\frac{\lambda}{\lambda^*}\right)^2 \left[1 - \left(1 - \left(\frac{\lambda}{\lambda^*}\right)^2\right)^{3/2} \right] & \text{for } \frac{\lambda}{\lambda^*} > 1 \\ \frac{2}{3} \left(\frac{\lambda}{\lambda^*}\right)^2 & \text{for } \frac{\lambda}{\lambda^*} < 1. \end{cases} \quad (16)$$

λ^* is the cut-off wavelength given by

$$\lambda^* = \frac{1}{K} \sqrt{2d/\rho} \quad , \quad (17)$$

where d is the guide width and ρ the radius of curvature. The dimensionless function $(\bar{\omega}/\omega_S)$, which is the ratio of the average solid angle for the bent guide to that of a straight guide of identical reflecting material is plotted in Fig. 3. Note that this depends only on (λ/λ^*) and so can be used for any reflecting material and any radius of curvature.

The above equations have been used, in combination with the source spectral distributions of Fig. 2, to calculate the intensities delivered by various beam tubes and guides. The results are shown in Fig. 4 and the associated angular divergences are shown in Fig. 5. Note that the cold guide curves are for the usual Ni coating and are based on the "cold A" curve of Fig. 2. These intensities and angular divergences can be converted to ^{58}Ni by multiplying by 1.4 and to the supermirror case by multiplying by 4.8.

B. Intensity at Sample - Crystal Monochromator

We start with the usual Cooper and Nathans probability function (horizontal part only).

$$P_H(\Delta\lambda, \gamma_1) = P_M \exp \left\{ -\frac{1}{2} \left[\left(\frac{\gamma_1 - (\Delta\lambda/\lambda) \tan \theta_M}{\eta_M} \right)^2 + \left(\frac{\gamma_1 - 2(\Delta\lambda/\lambda) \tan \theta_M}{\alpha_0} \right)^2 + \left(\frac{\gamma_1}{\alpha_1} \right)^2 \right] \right\}. \quad (18)$$

The intensity on the sample is given by

$$I(\lambda) = \phi''(\lambda) \delta_V \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} P_H(\Delta\lambda, \gamma_1) d\Delta\lambda d\gamma_1 \text{ [n cm}^{-2} \text{ sec}^{-1}] . \quad (19)$$

The vertical divergence for a flat monochromator is

$$\delta_V = \frac{h_S}{L_0 + L_1} , \quad (20)$$

where h_S is the source height, L_0 is the source to monochromator distance, and L_1 is the monochromator to sample distance. For a vertically focused monochromator⁷

$$\delta_V = \frac{h_M}{L_1} , \quad (21)$$

where h_M is the monochromator height. Performing the integrations in Eq. (19), we obtain

$$I(\lambda) = \phi''(\lambda) P_M(\lambda) \delta_V \lambda \cot\theta_M \frac{2\pi}{8\lambda n^2} \frac{\bar{\alpha}_0 \bar{\alpha}_1 \bar{\eta}_M}{\sqrt{\bar{\alpha}_0^2 + \bar{\alpha}_1^2 + 4\bar{\eta}_M^2}} , \quad (22)$$

where the width parameters have been converted to FWHM values,

$$\bar{\alpha}_i = 2\sqrt{2\lambda n^2} \alpha_i, \text{ etc.} \quad (23)$$

The factor $P_M(\lambda)$ is the peak absolute reflectivity of the crystal and the numerical factor $2\pi/8\lambda n^2$ is equal to 1.13. The FWHM in the wavelength distribution of neutrons incident on the sample, obtained by integrating Eq. (18) over γ_1 , is

$$\frac{\Delta\lambda}{\lambda} = \frac{1}{2} \frac{\Delta E}{E} = \cot\theta_M \left(\frac{\bar{\alpha}_0^2 \bar{\alpha}_1^2 + \bar{\alpha}_0^2 \bar{\eta}_M^2 + \bar{\alpha}_1^2 \bar{\eta}_M^2}{\bar{\alpha}_0^2 + \bar{\alpha}_1^2 + 4\bar{\eta}_M^2} \right)^{1/2} \quad (24)$$

Similarly the horizontal divergence (FWHM), obtained by integrating Eq. (18) over $\Delta\lambda$, is

$$\Delta\gamma_1 = \alpha_1 \left[\frac{\bar{\alpha}_0^2 + 4\bar{\eta}_M^2}{\bar{\alpha}_1^2 + \bar{\alpha}_0^2 + 4\bar{\eta}_M^2} \right]^{1/2}. \quad (25)$$

Note that for the special case $\alpha_0 = \alpha_1 = \eta_M$,

$$I(\lambda) = \phi''(\lambda) P_M(\lambda) \delta_V \lambda \cot\theta_M (1.13) \frac{\bar{\eta}_M^2}{\sqrt{6}} \quad (26)$$

$$\frac{\Delta\lambda}{\lambda} = \cot\theta_M \frac{\bar{\eta}_M}{\sqrt{2}} \quad (27)$$

$$\Delta\gamma_1 = \alpha_1 \sqrt{5/6}. \quad (28)$$

Another interesting case is $\alpha_0 \gg \alpha_1 = \eta_M$, for which

$$I(\lambda) = \phi''(\lambda) P_M(\lambda) \delta_V \lambda \cot\theta_M (1.13) \bar{\eta}_M^2 \quad (29)$$

$$\frac{\Delta\lambda}{\lambda} = \cot\theta_M \bar{\eta}_M \sqrt{2} \quad (30)$$

$$\Delta\gamma_1 = \bar{\alpha}_1. \quad (31)$$

In cases where Soller collimators are not used before the monochromator, the value of α_0 can be taken as the horizontal divergence of the tube or guide discussed previously.

III. COMPARISON WITH PULSED-SOURCE SPECTRA

The connection between $I(E)$ plotted in Fig. 1 of the notes by Crawford, Russell and Carpenter and $d^2\phi/d\Omega d\lambda$ plotted in my Fig. 2 is apparent by

observing that, in their notation,

$$\phi(E) = \frac{I(E)}{L^2} = \frac{I(E)}{A_{\text{Mod}}} \frac{A_{\text{Mod}}}{L^2} = \frac{I(E)}{A_{\text{Mod}}} \delta_H \delta_V .$$

So their quantity $I(E)/A_{\text{Mod}}$ is equivalent to my $d^2\phi/d\Omega dE$. Since they have used $A_{\text{Mod}} = 100 \text{ cm}^2$ the $I(E)$ values of their Fig. 1 should be multiplied by 10^{-2} to compare with my $d^2\phi/d\Omega dE$. The comparison with the time-averaged pulsed source spectra is shown in Fig. 6 after converting my wavelength distributions of Fig. 2 to energy distributions by multiplying by $\lambda/2E$.

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Fig. 1. Calculated thermal distribution compared to H12 beam tube, ILL.

Fig. 2. Estimated flux distributions for 5×10^{15} reactor.

Fig. 3. Bent guide cut-off function.

Fig. 4. Spectral distributions of various beams from a 5×10^{15} reactor.

Fig. 5. Angular divergence of tubes and guides of Fig. 4.

Fig. 6. Comparison of source spectra for 5×10^{15} reactor and state-of-the-art pulsed source. Note that for the pulsed source time-averaged, not peak, intensities are used.

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