1 of 2

Neutron Sources and Applications

Report of a Review held at
Oak Brook, Illinois

September 8–10, 1992

Edited by

D.L. Price
Argonne National Laboratory
Argonne, Illinois 60439

J.J. Rush
National Institute of Standards and Technology
Gaithersburg, Maryland 20899

MASTER

Sponsored by the
U.S. Department of Energy
Division of Materials Sciences
Office of Basic Energy Sciences

rp

Contents

1	Preface	1-1
2	Neutron Sources	
2.1	Reactor Sources	2-1
2.2	Pulsed Spallation Sources for Neutron Scattering.....	2-8
3	Neutron Scattering Applications	
3.1	Condensed Matter Physics	3-1
3.2	Materials Science	3-10
3.3	Chemistry	3-23
3.4	Polymers and Complex Fluids.....	3-32
3.5	Biology.....	3-42
3.6	Instrumentation.....	3-49
4	Other Neutron Applications	
4.1	Isotopes	4-1
4.2	Materials Irradiation.....	4-12
4.3	Materials Analysis and Imaging.....	4-23
4.4	Fundamental and Nuclear Physics.....	4-27
5	User Perspectives	
5.1	Perspective of the Neutron Scattering Society of America.....	5-1
5.2	University Education, Research, and Reactor Community Perspective.....	5-18
Appendix A: Advanced Neutron Source Performance Goals and Conceptual Design Parameters.....		A-1
Appendix B: Current and Projected Isotope Usage in the U.S.....		B-1
B.1	Isotopes in Common Usage.....	B-2
B.2	Current and Projected ²⁵² Cf Neutron Usage.....	B-3
B.3	Sales of Reactor-Formed Radioisotopes	B-4
B.4	Estimate of Future Demand for ⁸⁹ Sr.....	B-5
B.5	Estimated Annual U.S. Markets for Reactor-Formed Radioisotopes	B-5
Appendix C: Summaries of Reports on Related Uses of Neutron Sources		C-1
C.1	Positron Spectroscopy of Materials.....	C-2
C.2	Applications of Muons.....	C-3
Appendix D: List of Acronyms		D-1

Figures

2.1	Schedule and Construction Costs of the New JRR-3 Reactor	2-6
2.2	Staff Levels Associated with Construction and Commissioning of the New JRR-3 Reactor.....	2-7
2.3	Target Design with Flux-Trap Moderators and Shielding for the Spallation Neutron Source at LANSCE.....	2-19
2.4	Expanded Plan-View Schematic of the LANSCE Target/Moderator Arrangement.....	2-20
2.5	Schematic Layouts of Various Moderator Geometries	2-21
3.1	The Location of Pyridine in the Catalyst, Potassium Zeolite-L, Determined by Neutron Diffraction	3-26
3.2	Complementarity of a Reactor and a Pulsed Source.....	3-51
5.1	Age Distribution of the U.S. Neutron Community	5-7
5.2	Subject Matter of Papers in Past Five International Conferences on Neutron Scattering.....	5-19
5.3	Origin of Neutron Scattering Papers Presented during Past Five International Conferences on Neutron Scattering	5-20

Tables

2.1	Reactor Personnel and Operating Costs of the New JRR-3	2-5
2.2	Spallation Neutron Sources Operating or under Construction.....	2-12
2.3	Spallation Neutron Sources under Study	2-13
2.4	Existing Spallation Neutron Sources.....	2-14
2.5	Predicted 4π -Equivalent of Thermal Flux from the Moderator Surface for Various Target Systems	2-22
2.6	Parameters of Proposed LANL and ANL Accelerator Systems	2-24
3.1	Examples of Zeolite Structures Studied by Powder Neutron Diffraction	3-27
4.1	Medical Isotopes Produced via Neutron Reactions	4-5
4.2	Production and Use of Iridium 192.....	4-7

Tables (Cont.)

4.3	Summary of Reactors Currently Available in the U.S. for Controlled Materials Irradiations	4-17
5.1	Costs and Availability of Neutron Scattering Sources.....	5-9
5.2	Summary of User Statistics for FY 1991	5-11
5.3	European Major Facilities	5-11
5.4	European Neutron Scattering Facilities	5-13

1 Preface

The Panel on Neutron Sources, chaired by Professor Walter Kohn of the University of California, Santa Barbara, was set up in July 1992 by the Basic Energy Sciences Advisory Committee (BESAC) of the Department of Energy (DOE) at the request of Dr. William Happer, Director of the Office of Energy Research. It was asked to provide advice and guidance on the future of neutron sources and related scientific and technological applications in the United States. The Panel's report was published in January 1993 under the title *Neutron Sources for America's Future* (DOE Report ER-0576P).

In order to provide the Panel with a sense of the current thinking worldwide on the perceived needs for neutron sources of different types and the applications and capabilities of these sources, a Review of Neutron Sources and Applications was held at Oak Brook, Illinois, during September 8-10, 1992. This review involved some 70 national and international experts in different areas of neutron research, sources, and applications. Separate working groups were asked to: (1) review the current status of advanced research reactors and spallation sources; and (2) provide an update on scientific, technological, and medical applications, including neutron scattering research in a number of disciplines, isotope production, materials irradiation, and other important uses of neutron sources such as materials analysis and fundamental neutron physics.

This report summarizes the findings and conclusions of the different working groups involved in the review, and allows us to share with the community at large some of the best current expertise on neutron sources and applications.

We wish to thank the participants in the review, who gave their time and efforts on very short notice, and especially the chairmen of the working groups who were responsible for producing the written reports. We also acknowledge the thorough and painstaking efforts of Dr. Frank J. Rotella in editing the manuscript and the secretarial efforts of Barbara Salbego and Judith Robson in its production. At the time of editing these proceedings (March 1993), there have already been positive actions on neutron sources by the federal government. The President's budget request for FY 1994 contains a line-item obligation of \$26M as a first step towards construction of the ANS at Oak Ridge National Laboratory (ORNL), along with out-year projected spending of \$1.2B through FY 1998. The DOE budget also includes \$5M to initiate a design study for a 1-MW pulsed spallation source. If these actions hold up, the efforts of the participants will have been well spent.

D.L. Price and J.J. Rush
Chairmen
Review of Neutron Sources and Applications

2 Neutron Sources

2.1 Reactor Sources

P. Ageron (ILL, France)
 K. Boning (Technische Universität München, Germany)
 R.J. Budnitz (Future Resources Associates, Inc.)
 B. Farnoux (Saclay, France)
 G. Kinne (BNL), Chairman
 J.A. Lake (INEL)
 R.F. Lidstone (AECL, Canada)
 S. Matsuura (JAERI, Japan)
 C.D. West (ORNL)

2.1.1 The Advanced Neutron Source

The working group on reactor sources reviewed expected performance of the ANS against the design goals established by the National Steering Committee for the ANS. The project goals set by the representatives of national user communities that comprise NSCANS include the following principal requirements (fluxes in units of 10^{15} n/cm²·s):

	Scientific goals	ANS baseline performance
• <u>Neutron scattering</u>		
peak thermal flux in the reflector	5–10	7.4
thermal flux at cold sources	2–4	4
thermal flux at hot source	≥ 1	1
• <u>Transuranic production</u>		
epithermal flux	≥ 0.6	1.1
annual ²⁵² Cf production, g	≥ 1.5	1.7
annual ²⁵⁴ Es production, μg	≥ 40	60
• <u>Materials irradiation</u>		
fast flux in central facility	≥ 1.4	3.1
fast flux in reflector facility	≥ 0.5	0.4
• <u>Materials analysis</u>		
flux in pneumatic tubes PT1–3	> 0.2	0.3
flux in pneumatic tubes PT4–5	> 0.05	0.09
flux in hydraulic tube PF1	0.01–0.04	0.03
flux in hydraulic tube PF2	0.005–0.008	0.006

The baseline ANS design meets or exceeds these principal requirements. With regard to neutron scattering, it exceeds the peak-reflector thermal-flux goal by almost 50% and the minimum cold-flux goal by 100%. With regard to isotope production, it exceeds the californium goal by 13% and the einsteinium goal by 50%. With regard to materials irradiation testing, it exceeds the central-facility-flux goal by 120%. With regard to materials analysis it exceeds the pneumatic-tube minimum fluxes by 50 to 100% and the hydraulic-tube minimum fluxes by 20 to 200%. Overall, considering the detailed set of scientific user goals, the baseline ANS design meets essentially all minimum requirements, and in most cases significantly exceeds minimum requirements. A complete list of performance goals and conceptual design parameters is given as Appendix A.

Importantly, the baseline designs provide a complete "user-friendly" facility, including experimental halls, laboratory and sample preparation areas, office areas, computer-analysis services, and guest housing.

The basic design concept for the ANS relies on technologies and design approaches that have already been proven at existing reactors. Examples include involute fuel plate geometry and cermet fuel meat, as used at HFIR, uranium silicide fuel from the RERTR program, and a cold source modeled from experience at the Institut Laue-Langevin (ILL). However, in a few important areas the design concept goes somewhat beyond what has been demonstrated. Therefore, several series of experiments, tests, and engineering analyses have been planned to develop the needed engineering information or to confirm that the design margins are adequate. Some of these tests and analyses are already underway, and a few have been completed. Although surprises that would invalidate the fundamental design concept are always possible, we believe that they are unlikely. We are confident that the planned program of confirmatory tests and experiments will amply demonstrate that the facility will meet present and foreseeable safety requirements with adequate margin.

The ANS design team has taken great pains to understand the full range of present and foreseeable requirements in the areas of public health and safety, occupational safety, and environmental protection. The design concept has carefully considered these and has attempted to assure that all such requirements will be met. We have been impressed both with the comprehensive approach taken, including the way advice about the various requirements has been sought out, and with the apparent success that the current design has achieved so far in this regard. Especially in the reactor safety area, we believe that the ANS is highly likely to pass successfully the very stringent regulatory reviews that will inevitably occur over the next several years on the part of the DOE and perhaps other groups.

The reactor-sources working group judges the ANS project cost estimate to be comprehensive, thorough, and conservative (i.e., it is perhaps an overestimate). So-called "bottom-up" labor estimates were developed for project participants based on their individual tasks. The construction estimate is based on an extensive database that incorporates detailed physical quantity takeoffs, budget estimates from equipment suppliers, experienced judgment, past project history, industry averages, concept-engineering input, and HFIR experience. The engineering estimate is based on system-by-system documentation that includes a complete set of design requirements and descriptions plus pertinent system drawings. The specifications for procuring

equipment and materials are based on commercial standards for the nuclear power industry. In particular, the design and cost already account for full compliance with all current and foreseeable safety and environmental regulations and orders. Considering the extent of reliance on existing technology, an appropriate contingency (averaging 20.6% for line items) has been applied reasonably to cover costs that might result from incomplete design, unforeseen and unpredictable conditions (e.g., major changes in anticipated regulatory requirements), and uncertainties in the defined scope of work.

The reactor-sources working group emphasizes that the ANS baseline design provides a complete user-oriented facility for neutron scattering, isotope production, and materials irradiation testing and analysis. The project scope includes not only the neutron source (\$458M), but also the experimental infrastructure (\$434M for experiment systems, cold sources, beam halls, laboratory space, support, etc.), and operational-support facilities (\$502M for water and electrical supplies, a detritiation facility, plant instrumentation and control systems, the operational support building, guest accommodations and other structures, QA and environmental services, and project administration). It should be recognized that most of these project elements beyond the neutron source will be required for any facility or combination of facilities that fully meets the set of scientific requirements established by the NSCANS.

The line item cost of \$1700M (1992 dollars) covers a facility that exceeds nearly all of the many scientific requirements. It is perhaps worth noting that more than 50 separate specifications for flux, spectrum, heating rates, number of beams, and analytical capabilities have been provided by the many scientific communities involved, who mainly articulate their needs through a strong and diverse NSCANS. These requirements are carefully documented and tracked by the project.

In addition to the line item cost, the total project cost, as now defined by the DOE, includes operational costs (e.g., for operator training, fuel inventory, and R&D) incurred prior to actual operation. These total \$399M (1992 dollars).

Since the cost estimate was prepared in June of 1992, value engineering studies by the project have identified about \$75M of cost reductions, mostly rearrangements of building and site layouts, that could be implemented without loss of scientific utility. We endorse these changes.

Further reductions in cost can be achieved through design and scope changes, such as reducing the power from 330 MW to 200–220 MW, which would still meet the minimum scientific requirements and would raise the possibility of shortening the time to full operation by as much as two years: the NSCANS is evaluating the desirability of such a change.

The projected operating costs for the ANS are approximately \$80M per year. This includes the costs for reactor operations, management and administrative support, maintenance, QA and health and safety support, training, utility and equipment costs, and reload cores. The operating staff numbers 388. The working group believes this cost estimate to be inclusive of all the major needs for reactor facility operations. The ANS estimate was compared with the current operating budgets for the HFIR and HFBR reactors. The proposed ANS staff size and annual operating cost

are higher than the total of HFIR and HFBR combined. The ANS staff believe this to be reasonable based on the increased staff support required to support the larger total number of research facilities at the ANS, the detritiation plant, and the electrical requirements for the ANS cryogenic sources. The working group recommends that ANS evaluate further the requirements for reactor technical support management and administration in steady-state operations. Overall, we believe that the ANS operating cost estimate is comprehensive and may be conservatively high.

2.1.1.1 The Short Term

The working group, supported by the thoroughness and depth of the ANS conceptual design report, judges that the project is ready *now* to move into a line item stage in accordance with the schedule laid out in that report. We recommend, for FY 1994, the project's request of \$42.5M in line item funding and \$16.1M in expense and capital equipment money.

To minimize delays if the project is not a line item, at least \$43M of operating funds are needed in FY 1994. Less than this level of funding would mean delays of up to one year, and we note that this scientific facility is urgently needed, and also that each year of delay has added, and will continue to add, approximately \$75M to the total project cost.

One of the key challenges that the ANS design team has faced since the project's inception is that the DOE criteria by which the design will be judged, especially in the safety arena, are not yet known in detail. To compensate for this, the design team has attempted to meet what it considers to be a set of safety criteria roughly equivalent to those embedded in the Nuclear Regulatory Commission's (NRC's) requirements for large power reactors, subject area by subject area. We believe that the development by the DOE of a complete set of criteria by which the ANS will be judged, especially in the safety arena, is of urgent priority. Furthermore, the review process is still unknown, and the DOE organization that will review the ANS design has yet to be established and staffed. We urge the DOE to act very soon on these very important issues so that the ANS can be kept on schedule, thereby avoiding cost escalation.

The ANS design is based on relatively modest extrapolations of well-proven, plate-fueled, water-cooled research reactor technology. The ANS performance therefore does not represent the ultimate that could be achieved at the limits of reactor technology. As an example, the working group heard a presentation on scoping studies that are being performed to explore the potential performance capabilities of a water-cooled, packed-particle bed reactor. The packed-particle bed reactor may be able to operate at very high power density in a compact core, and thereby generate neutron flux levels several times higher than those achieved by the current ANS design. The packed-particle bed reactor R&D effort is currently being supported by the U.S. Air Force Space Nuclear Thermal Propulsion program. Considerable additional detailed design and development would be required for application of this concept to produce a reliable reactor neutron source. Other examples of advanced concepts are also known to our group.

2.1.2 Research Reactors in Germany

There are currently four major research reactors in operation in Germany: Munich, Jülich, Geesthacht, and Berlin. During about the last decade, one of these reactors (Berlin) has been basically renewed and upgraded, and two others (Jülich and Geesthacht) have received major improvements of their experimental facilities — in particular, cold sources and neutron guide halls.

It is planned to replace the oldest research reactor — the FRM at Munich — by a totally new facility. This new research reactor, the FRM-II, will be a small but high performance national neutron source featuring a very compact, light-water cooled core in the center of a large heavy water tank, the maximum thermal neutron flux being calculated as 8×10^{14} n/cm²·s at 20 MW power. This university-operated source has been optimized with respect to neutron scattering applications but is nevertheless a multipurpose facility. In the field of neutron scattering, it will be used to train students, develop experimental techniques, and perform all those experiments which do not need to use the larger international source at Grenoble, France (ILL). The total cost of the project has been established as slightly below \$400M (in 1992 dollars, including taxes); this figure also includes the construction of the cold source, irradiation facilities, and about 10 scattering instruments. The present project status is that the conceptual design and the safety report have been completed so that the application for nuclear licensing of the FRM-II can be submitted immediately after a funding decision.

2.1.3 JRR-3

Details of the schedule, costs, and staff levels associated with construction and commissioning of the new JRR-3 reactor at the Japan Atomic Energy Research Institute (JAERI) at Tokaimura, Japan, are provided in Table 2.1 and Figs. 2.1 and 2.2.

TABLE 2.1 Reactor Personnel and Annual Operating Costs of the New JRR-3

<u>Reactor Personnel</u>	
Reactor Operation and Maintenance	33
Planning and Core Management	6
Fuel Management	6
Operations and Maintenance of Utilization Facility	15
Radiation Protection	5
Water, Gas, and Electricity Control	10
<u>Annual Operating Costs (\$)</u>	
Fuel	4M (1.6 cores)
Salary	50k (per person)
Other (Total)	14M (8 cycles)

FIGURE 2.1 Schedule and Construction Costs of the New JRR-3 Reactor.

FIGURE 2.2 Staff Levels Associated with Construction and Commissioning of the New JRR-3 Reactor.

2.2 Pulsed Spallation Sources for Neutron Scattering

G.S. Bauer (PSI, Switzerland)
J.M. Carpenter (ANL)
M. Craddock (TRIUMF, Canada)
D. Dowd (CEBAF)
H.A. Grunder (CEBAF), Chairman
G. Lambertson (LBL)
G. Rees (RAL, U.K.)
G.J. Russell (LANL)
B. Weng (BNL)

2.2.1 Introduction

The following report deals with accelerator-based, short-pulsed spallation neutron sources for slow neutron scattering applications. We distinguish these from other types of accelerator-driven sources, namely steady-state sources and long-pulse, "time-modulated" sources. We deal with facilities that are affordable in the context of neutron scattering research and achievable with present-day technology or modest extensions of it. Furthermore, the installations we deal with here are of the scale of central national or international research institutes and are comparable in their performance with very high flux reactors. Thus, though it might be useful and informative to take a broader or longer-term view, we do not deal with spallation sources capable of producing time-average neutron fluxes equal to those provided in the highest-flux reactor concepts or with facilities that provide large volumes at high flux. Neither do we deal with small sources appropriate for local or special purposes.

Steady developments in the accelerator field have led to the present situation in which beams of protons of several GeV energy, with time-average power in the range of several megawatts, short-pulse time structure, and low repetition frequency, are accessible. In the past decade or so, results from pulsed sources based on accelerator beams of lower power have demonstrated the effectiveness of a wide variety of instruments based on these sources, and they provide a highly encouraging basis for extrapolation of their performance to higher intensities. At the same time, however, it must be recognized that the techniques for utilization of the pulsed sources have not reached the same level of maturity as those for using steady, reactor-produced beams. In view of these observations, comparing the scientific usefulness of steady, reactor-produced neutron beams with pulsed, accelerator-produced beams remains a difficult problem because the comparison must be made either in very detailed terms (i.e., instrument-by-instrument and purpose-by-purpose) or in very general terms, with sweeping arguments and general averages hard for all but the wisest generalist to formulate or appreciate. In spite of these difficulties, this working group has done its best to characterize the prospects for pulsed spallation neutron sources and to lay the basis for comparison with the steady reactor sources.

2.2.2 Key Features of a Pulsed Spallation Neutron Source

The total number of neutrons produced in a pulsed spallation neutron source (PSNS) is relatively small compared to a high-flux research reactor. The volume in which the neutron flux is available is only of the order of a few liters. As a consequence, certain applications such as isotope production, high-flux materials irradiation, or neutron activation cannot be readily accommodated on a pulsed source designed for neutron scattering and would not benefit from the time structure. These applications are therefore excluded in the following discussion.

Owing to the fact that the proton beam driving the neutron production can be delivered in short pulses of less than one microsecond, a peak-to-average flux ratio of order 1000 can be obtained. The large variety of different instruments and methods used in neutron scattering to arrive at the desired information makes it difficult to quantify the gain resulting from the exploitation of the time structure. However, based on the experience with existing sources and on a detailed analysis carried out for the long-pulse (250 μ s) version of the German SNQ project, an overall enhancement factor of 100 appears an appropriate number for a PSNS. (The enhancement factor is the factor by which the flux of a steady-state neutron source would have to be higher than the time-averaged flux of a pulsed source to give a comparable overall performance.) This does not, of course, account for a number of other advantages of spallation neutron sources as discussed below.

The high scientific potential of a PSNS results from the fact that, through its very properties, it allows use of neutrons in different ways and in a more efficient way than a steady-state source of any kind.

The facts that neutrons are generated in short pulses at times when no detection takes place and that different neutron energies arrive at the sample at different times result in the following advantages:

- Good signal-to-background ratio in most types of neutron scattering instruments and hence a possibility to observe more subtle effects with less effort;
- High accessible range of incident energies resulting in a wider range of energy and momentum transfer, which allows more complex systems to be investigated; and
- High intrinsic resolution in incident energy resulting in high precision data, allowing important details in the scattering pattern to be resolved.

- Spectrometer geometries can be fixed (as opposed to scans by moving parts of spectrometers at steady-state sources). This leads to:
 - Better utilization of the solid angle around the sample and hence increased data collection efficiency;
 - Easier use of complex sample environment equipment to keep the specimens in desired states (pressure, temperature, magnetic field, etc.); and
 - The possibility of taking data with different information content simultaneously (e.g., structural and dynamic information by simultaneous diffraction and spectroscopy; structure and particle size or density fluctuations by diffraction and small-angle scattering, etc.). This can be extremely valuable in the study of transitional states.
- Finally, the pulsed nature of the source greatly facilitates the study of time-dependent phenomena, either by phasing the imposed starting conditions of the sample to the source pulse or by using several source pulses to measure the same quantities while the state of the sample is changing.

The features inherently present in a PSNS can be generated in a steady-state source but at the expense of substantial loss factors.

- Energy separation by monochromating crystals results in only one energy band being usable throughout the whole time, whereas in a PSNS, a large number of energy bands can be used between pulses to obtain the same or additional valuable information.
- Beam chopping will only produce pulses at less than the time-averaged flux level, suppressing the source neutrons for more than 99% of the time and leading to less favorable conditions with respect to resolution.
- Periodically operating devices such as Doppler-shift crystals can operate in phase with a PSNS and hence use the full time-averaged flux, whereas on steady-state sources, the required conditions only prevail for a fraction of the time.

Other important factors in the comparison between a state-of-the-art steady-state reactor and a state-of-the-art PSNS include:

- A PSNS can be constructed at lower cost for a comparable level of performance and is very cost-effective per "useful" neutron.
- A PSNS has a high development potential in proton beam parameters, number of target stations served by one accelerator system, and on the instruments' side.
- A PSNS allows more freedom in designing or modifying the source according to the users' needs due to the absence of retro-effects of the moderators on the target yield.
- A PSNS does not produce transuranic waste and virtually none of the hazardous fission products.
- There is no need to use or handle proliferation-prone materials in a PSNS.
- There is an inherently lower risk associated with a PSNS since it does not involve exothermic or chain reactions.

If technical applications of very high-power accelerators such as Accelerator Production of Tritium (APT), Accelerator Transformation of Waste (ATW), or driven subcritical nuclear power plants are to be developed or seriously studied, an intermediate facility in the 1- to 5-MW range as considered for the next generation PSNS would be a logical and necessary step. An overview of the parameters of high-power pulsed spallation neutron sources under study is given in Tables 2.2 and 2.3. It can be seen that, with an enhancement factor of about 100, the effective flux of pulsed spallation neutron sources can fall well within the regime of a high-flux reactor ($\sim 10^{15}$ n/cm²·s).

2.2.3 State of the Art for Spallation Neutron Sources

2.2.3.1 Introduction

Pulsed sources for neutron scattering research were originally based on an electron linear accelerator and a high-power target station. The situation changed in the 1970s when the advantages of proton-based spallation for reduced target heating were pointed out at Argonne National Laboratory (ANL). Subsequently, ANL pioneered the first spallation source.

TABLE 2.2 Spallation Neutron Sources Operating or Under Construction

Facility Country	Proton Energy (MeV)	Nominal Proton Current (μ A)	Nominal Beam Power (kW)	Source Pulse- Width (μ s)	Pulse Repetition Rate (Hz)	Target	Time-Averaged Thermal Flux ^a (n/cm ² ·s)	Peak/ Average Flux ^{b,c}	Year
KENS (Japan)	500	6	3	0.1	20	²³⁸ U	7×10^{11}	2500	1980
IPNS (U.S.)	450	15	7	0.1	30	²³⁸ U	1.4×10^{11}	1700	1981
						²³⁵ U	3.5×10^{11}	1700	1988
LANSCE ^{d,e} (U.S.)	800	75	60	0.25	20	W	8.5×10^{11}	2500	1977
							1.1×10^{12}	2500	
ISIS ^e (U.K.)	800	150	120	0.27	50	Ta	1.1×10^{12}	1000	1985
						²³⁸ U	2.3×10^{12}	1000	
SINQ (Switzerland)	575	1000	575	continuous	continuous	under study	1×10^{14}	1	1995

^a 4π -equivalent of differential useful thermal neutron current from a moderator surface.

^b Scaled from ISIS according to Monte Carlo calculations, by G.J. Russell.

^c Performance estimates are based on very preliminary measurements and theoretical extrapolation. There is some disagreement between reported results of measurements and calculations of scaling as represented in this table. However, these values are roughly indicative of performance of the various facilities.

^d The two values for the LANSCE performance correspond to 10-cm $\times$ 10-cm and 12-cm $\times$ 12-cm moderator fields-of-view which are in use.

^e Measured values for ISIS and LANSCE may be low by as much as 20%.

TABLE 2.3 Spallation Neutron Sources under Study

Facility Country	Proton Energy (MeV)	Beam Power (kW)	Source Pulse- Width (μ s)	Pulse Repetition Rate (Hz)	Time- Averaged Flux (n/cm ² ·s)	Peak/ Average Flux
KENS-II (Japan)	1000	100	0.2	50	3×10^{12}	
AUSTRON (Austria)	1600	200	<1	25	5×10^{12}	
ESS* (Europe)	0.8–3.0 (2nd target)	4000	<1	40	5×10^{13}	1200
		1000	<1	10	1.5×10^{14}	800
IPNS-II (U.S.)	1500	750	0.5	30	1.3×10^{13}	1750
LANSCE-II (U.S.)	800	1000	0.5	60	1.6×10^{13}	800

- * Two target stations: (1) High power, decoupled moderator.
(2) Low power, coupled moderator.

Four such sources are now in existence, three based on an H⁻ linac and a proton synchrotron, and one on a H⁻ linac and a proton compressor ring. All four make use of charge exchange injection to strip the H⁻ beam to protons and allow a large accumulation of proton beam current. The characteristics of the accelerators and targets for these sources are given in Table 2.4.

2.2.3.2 Limitations of Existing Spallation Neutron Sources

The four existing sources have little potential for increasing their neutron production without major modifications. IPNS has introduced an enriched uranium target that increases intensity by 2.5. ISIS is considering extending its neutron scattering program by the addition of a new 10-Hz target, together with a range of new instruments. Los Alamos National Laboratory (LANL) has proposed a LANSCE upgrade to increase the number of neutron beam lines serviced by ~33 % (12 to 16). This would be accomplished by adding two new moderators.

TABLE 2.4 Existing Spallation Neutron Sources

Source	H ⁻ Linac Energy (MeV)	Ring Energy (MeV)	Average Beam Power (kW)	Repetition Rate (Hz)	Target
KENS (Japan)	40	40–500	2	20	²³⁸ U
IPNS (U.S.)	50	50–450	7	30	²³⁸ U
ISIS (U.K.)	70	70–800	120	50	²³⁸ U, Ta
LANSCÉ (U.S.)	800	800	60	20	W

The various limitations of the existing accelerator sources are:

1. the available H⁻ ion source currents;
2. the level of transverse space charge forces in the 3 synchrotrons;
3. the level of longitudinal space charge forces in the LANSCE compressor ring (PSR);
4. the associated electron-proton instability observed in the PSR;
5. the resistive wall instability (head-tail) observed in the 3 synchrotrons (though all have found solutions for avoiding the effect);
6. the incomplete stripping of the H⁻ beam at the time of injection into the ring (e.g., approximately 2% of the beam remains as H⁰);
7. the subsequent stripping of a fraction of the H⁰ beam (that is in an excited quantum state) before removal from the ring, a phenomenon recently discovered at LANL;

8. the inelastic and elastic scattering, and energy loss, that occurs as the H^- particles and protons pass through the stripping foil;
9. longitudinal trapping losses in the 3 synchrotrons; and
10. resonance crossing due to the transverse space charge forces.

In the design of the next generation sources, these various limitations have to be addressed in Conceptual Design Reports. It is expected that these various issues can be successfully addressed with modest R&D efforts.

2.2.3.3 State-of-the-Art Technical Features for Next Generation Spallation Neutron Source

State of the art for some of the key technical features of the sources are:

1. 50–60-mA (max) dc H^- ion source currents;
2. input and output matching for high radio-frequency quadrupole (RFQ) linac transmission;
3. chopping and energy ramping of the H^- beam;
4. funneling of 2 low-energy linac beams into 1 main linac;
5. very reliable stripping foils with two unsupported "free" edges;
6. "optimized" H^- injection painting in (x, p_x, y, p_y, z, p_z) space;
7. reliable ceramic chambers for pulsed magnet vacuum chambers;
8. radio-frequency (rf) shields for the ceramic chambers, to reduce the coupling impedance between the beam and its environment;
9. "fast" harmonic corrections for quadrupole focusing fields;

10. sophisticated beam loss histogram displays;
11. efficient localized collection of lost beam;
12. magnet lattices for the rings which are designed around the constraints of an optimized H^- injection system.
13. the use of rf feedback and feedforward techniques in the radio-frequency systems of the rings.

The high-power targets that are used in the existing facilities include:

1. 0.25-MW (including target fission energy) depleted uranium target (ISIS); this target has a limited lifetime;
2. 0.17-MW tantalum target (ISIS);
3. 0.10-MW split tungsten target (LANSCE);

In addition, two low-power targets are in operation:

1. a special enriched uranium booster target (IPNS); and
2. a high-efficiency, low-power target (KENS).

The parameter extrapolations from the existing to the proposed new sources (in the U.S. and in Europe) are approximately :

	Existing	Short-Term	Long-Term
Average Beam Power	0.15	1	5 MW
Maximum Protons per Pulse	4.0	10	$20 (\times 3) \times 10^{13}$

Here the factor ($\times 3$) is related to the scheme of using 3 compressor rings in one of the proposals. The following sections of the report discuss the feasibility, and costs involved, for these parameter extrapolations.

2.2.4 Target Systems for Pulsed Spallation Neutron Sources

Operating experience at existing pulsed spallation neutron sources has demonstrated that spallation targets, moderators, and reflectors can operate with acceptable lifetimes at proton beam powers of 0.12 MW. A target system for a 1-MW pulsed spallation neutron source is readily achievable using present technology. A 5-MW pulsed spallation neutron source is feasible; however, some work must be done to validate the concept.

2.2.4.1 Introduction

The *target system* for PSNS's must provide spectrometers with potent neutron beams of appropriate time-structure and energy-spectral distributions. The target system is an ensemble of components (target, moderators, poison/decoupler/liners, and reflectors) which work in concert to produce proper neutron pulses. A typical target system is illustrated in Fig. 2.3; the target system is surrounded by a biological shield.

Energetic neutrons (2–3 MeV) are produced by the spallation process when the target is struck by high-energy protons from the accelerator. Moderators slow down source neutrons from the target to lower (useful) energies. Neutrons leaking from moderators into flight paths are used for scientific research. To keep the neutron pulses as short as possible, only hydrogen-containing materials are used as moderators in PSNS's. Poison, decouplers, and liners (see Fig. 2.4) are used to isolate the moderators neutronicly and tailor the intensity and pulse width of the leakage neutrons. Reflectors enhance neutron intensities from moderators by allowing neutrons multiple chances to slow down inside the moderators and leak as useful neutrons. The reflector can also act as part of the overall neutron production system by (n , $2n$) and spallation reactions.

The geometric arrangement of targets and moderators can be optimized to enhance the neutronic performance of a target system. Some typical geometries are illustrated in Fig. 2.5. The neutronic performance of a spallation target system must be optimized by including the synergism of *all* components (including geometry) of the target system.

2.2.4.2 Operating Experience at Existing Spallation Neutron Sources

As mentioned previously, there are four operating PSNS's: KENS, IPNS, LANSCE, and ISIS. Collectively, the operation of these facilities has demonstrated the following performances for the components of a PSNS:

- Target Materials:
 - Tantalum and tungsten targets have operated successfully with no problems. Uranium has exhibited a marginally acceptable lifetime (< 6 months) at a proton beam power of ~ 0.1 MW.
- Moderator materials:
 - Light water, no problems at proton beam powers of 0.12 MW;
 - Liquid hydrogen, no problems at proton beam powers of 0.12 MW;
 - Liquid methane, radiation damage has been observed but is controllable at a proton beam power of 0.12 MW;
 - Solid methane, exceptionally good moderator, but thermochemically unstable due to radiation damage at proton beam powers of 0.007 MW.
- Poisons/Decoupler/Liner materials:
 - Gadolinium, cadmium, and boral used successfully at proton beam powers of 0.12 MW.
- Reflector materials:
 - Beryllium has been used successfully as a reflector material.

The choice of the target-moderator-reflector arrangement has a profound effect on the parameters of the neutron beams available to the experimentalist. Therefore, beams suitable to a wide range of experiments can be provided on one, or even better, on two target stations driven by the same accelerator. Table 2.5, following Figs. 2.3-2.5, is taken from the report of a target technology workshop recently held at the Paul Scherrer Institute (PSI-Proceedings 92-03). It shows how pulse width (and hence, beam intensity) can be affected by different choices of materials. In this way, high-intensity or high-resolution beams with different spectral properties can be provided.

FIGURE 2.3 Target Design with Flux-Trap Moderators and Shielding for the Spallation Neutron Source at LANSCE.

FIGURE 2.4 Expanded Plan-View Schematic of the LANSCE Target/Moderator Arrangement. The Cold Moderator is Depicted on the Right Side.

FIGURE 2.5 Schematic Layouts of Various Moderator Geometries.

TABLE 2.5 Predicted 4π -Equivalent of Thermal Flux from the Moderator Surface for Various Target Systems (Moderators in Wing Geometry)

Moderator/ Reflector Configuration		$\phi \ 4\pi_{th}^a$	Reference	$\tau \ (\mu s)$	Temperature
1	Poisoned Decoupled H ₂ O	0.12	b	20	Ambient
2	Decoupled H ₂ O	0.40	b	60	Ambient
3	Coupled H ₂ O	1.33	c	140	Ambient
4	Decoupled liquid H ₂	0.37	b	120	20K
5	Coupled liquid H ₂	1.71	d	300	20K
6	Coupled H ₂ O/Lead Reflector	1.38	f	100	Ambient
7	Coupled liquid H ₂ /D ₂ O Reflector	0.78	e	710	20K
8	Coupled liquid H ₂ /Graphite Reflector	0.62	e	350	20K
9	Coupled liquid H ₂ /Lead Reflector	0.50	e	230	20K

^a Units are 10^{14} n/cm²·s per 1 MW of beam power at 1 GeV.

^b Scaled from ISIS performance.

^c CERN coupling, ISIS scaling.

^d KENS coupling, ISIS scaling.

^e Scaled from SNQ mockup at SIN.

^f Scaled from SNQ mockup at SATURNE.

2.2.5 Accelerator System for a 1-MW Source

2.2.5.1 Guidelines and Specifications

- a) The neutron flux is essentially proportional to the average power of the proton beam on the target. This unperturbed, steady-state thermal flux ($E < 0.625$ eV) for a spallation neutron source is 10^{14} n/cm²·s·MW if moderators are coupled (long pulses) or 10^{13} n/cm²·s·MW if moderators are decoupled and poisoned (short pulses).
- b) Full advantage of the time structure of a pulsed source requires a pulse duration less than a few microseconds and 10–60-Hz repetition rate.

- c) Following these guidelines, a selection of possible parameters for beam on target are:

proton energy	~1 GeV
average current	~1 mA
pulse length	~1 μ s
repetition rate	10–60 Hz
protons per pulse	$1-6 \times 10^{14}$

2.2.5.2 Approaches

In a pulse of a few microseconds duration having 10^{14} particles, the disruptive forces from space charge are strong. For that reason, a pulse cycle starts with the acceleration in a linac of a longer pulse of about 30 mA of H^- ions. Then at a moderately high energy, this charge is accumulated in a circular ring of a few microseconds orbit period. Injection employs stripping from H^- to H^+ and requires ~500 turns. The ring is designed to provide stable circulation of a beam at this energy of the required current and total pulse charge. Ejection to the target occurs in a single turn.

Presentations to the working group proposed two types of ring accumulator. The first of these is a dc storage ring where the beam is accelerated to full energy before injection. This can also be called a compressor ring, its purpose being merely to assemble the charges into a short pulse. In the second scheme, beam is accelerated to perhaps only 1/4 of its final energy and injected into a rapid-cycling synchrotron (RCS), where it is accelerated to final energy.

The working group also noted the approach that employs a linear induction accelerator (LIA). In this case, the ion source launches a pulse of about 5 amperes of protons. During the following acceleration in the LIA, this charge is compressed to the required 100 A and 1 μ s. From the LIA it is transported directly to the target. Additionally, the subpanel acknowledged superconducting technology as an option. Little information is available on superconducting linacs in the range of 1 GeV. The reader should note that a high efficiency in the rf-to-beam-power ratio forces the designer to relatively low gradients resulting in a long linac, and hence, appreciable capital costs. The question arises whether superconducting technology ($\times 3-6$ gradient) could shorten the linac with equally high or better rf power to beam power conversion.

Examples of accelerator systems were presented to the working group by LANL and ANL. The parameters of these are listed in Table 2.6.

TABLE 2.6 Parameters of Proposed LANL and ANL Accelerator Systems

	LANL	ANL
Beam Power	1 MW	0.75 MW
Repetition Rate	75 Hz	30 Hz
H ⁻ injector	30 mA, 75 keV 1 ms	50 mA 0.35 ms
RFQ linac	400 MHz, 2.5 MeV	not stated
DT linac(s)	400 MHz, 20 MeV 800 MHz, 100 MeV	425 MHz, 70 MeV
CC linac	800 MHz, 800 MeV	1,775 MHz, 400 MeV
Ring	compressor 10 ¹⁴ protons per pulse 0.8 GeV 0.45 μ s pulse	RCS 10 ¹⁴ protons per pulse 1.5 GeV 0.5 μ s pulse
Targets	Two (60 and 10 Hz)	Two (30 and 10 Hz)

Costs for these systems were used in arriving at the project costs that are presented in the working group's summary statement.

2.2.5.3 Issues and Outlook

The component units of these systems are based on present technology, with a minor increase in the charge storage capacity of the accumulator rings. Comments on the status and requirements of these elements follow.

Sources for H⁻ ions are operating in the range of 30–60 mA. Accelerated current of 30 mA has been demonstrated using an RFQ linac. The linac train is not at its performance limit, but careful emittance matching and chopping must be done in the low-energy stages to avoid radioactivation from losses at higher energies. Feedforward control can be applied to compensate for beam-loading transients in the rf systems.

A carbon or Al₂O₃ foil is used to strip H⁻ into H⁺ upon injection into the ring. Loss at this point must be limited to 10⁻⁴. A foil with an unsupported corner edge is desired to keep losses low

while placing successive turns into a uniform pattern within the acceptance phase space (called "painting"). Development of such foils to give longer operating life is in progress. Some H^0 particles in excited states can enter the ring and cause activation. These aspects of injection will benefit from R&D efforts.

Experience from ISIS can be applied in designing the ring lattice. A large linear acceptance is desired, and minimizing errors that drive betatron resonances will increase the tolerance for space-charge tune shifts. Excitations by a resistive-wall impedance may require feedback stabilization. Ejection is by means of a full-aperture kicker.

The requirements of the accumulator ring are more readily met if it is a static compressor type rather than rapid cycling. However, linac cost is reduced by injection at lower energy into the RCS. Such optimization and design choices must be addressed in a detailed design study.

2.2.6 Target System for a 1-MW Pulsed Spallation Neutron Source

A target system for a 1-MW pulsed spallation neutron source is readily achievable using present technology.

It is safe to assume that little development will be required to provide a water-cooled tantalum target and light-water and liquid hydrogen moderators for 1-MW proton beams. We can make this statement based on: a) operating experiences at both the ISIS and LANSCE PSNS's; b) the design study for the SINQ continuous spallation source; and c) operating experience at the LAMPF beam stop.

The ISIS facility at the Rutherford Appleton Laboratory is operating routinely at 0.12 MW of 800-MeV protons. Experience with a tantalum target indicates that such a target will pose no problems for a proton beam power of 1 MW. For uniform proton beam spot diameters of 7–9 cm, the peak power density in the tantalum target would be in the range of 0.8 to 1.3 MW/L which poses no insurmountable cooling problem. Light water and liquid hydrogen moderators are used routinely at both ISIS and LANSCE, and will pose no problems for use at a 1-MW source.

The SINQ (1.6 MW, 600 MeV) continuous spallation neutron source under construction at the PSI in Switzerland in many senses represents a model for a 1-MW spallation source. The SINQ source is slated for commissioning in 1995.

At LANL, the LAMPF beam stop is essentially a quasicontinuous (120-Hz) spallation neutron source running at 0.7–0.8 MW. Though the "target" material for the LAMPF beam stop is copper, the proton beam window and other beam stop structural materials provide valuable operation experience for radiation damage to materials exposed to a proton beam of about 1 MW. For example, the LAMPF inconel window is routinely subject to an incident proton flux of

$30 \mu\text{A}/\text{cm}^2$ and is still functioning satisfactorily after receiving a proton fluence of $1.3 \times 10^{22} \text{ p}/\text{cm}^2$. Similar experience exists at the PSI with aluminum windows. A 1-MW, 800-MeV PSNS would have proton current densities in the range of $15\text{--}25 \mu\text{A}/\text{cm}^2$; a proton fluence of $1.3 \times 10^{22} \text{ p}/\text{cm}^2$ would correspond to about 2 years of continuous operation, which would be a satisfactory lifetime.

A typical 1-MW PSNS would have two separate target areas. Such a scheme is included in the proposals by both LANL and ANL. In these proposals, one (high-power) target area would accept 75-80% of the 1-MW beam and operate at 30-60 Hz. The second (low-power) target area would receive the remaining proton beam and operate in the range of 10-15 Hz. Each target area would have its own *target system* suitably optimized to serve a certain class of instruments. Within a *target system*, the neutronic performance of individual moderators can be adjusted to satisfy experimental requirements without altering the target neutron production (unlike a reactor where such changes alter the reactivity of the core). In a PSNS with two target areas, some ~40 neutron beam lines would be served simultaneously by individual, neutronically optimized moderators. The number of beams that can be accommodated depends primarily on the number of moderators used and on the sector space available at the target shield (i.e., not used for proton beam transport, service areas, or handling cells). Examples exist for horizontal beam injection (KENS, IPNS, and ISIS) and vertical injection from the top (LANSCE) and from underneath (SINQ, under construction). Although vertical injection, in principle allows 2π of the circumference of the shield to be used, it has its problems too, and may become increasingly more difficult as the proton beam power increases. It is, however, feasible at 1 MW. In two cases (Moscow Meson Factory and KENS-II) it is even foreseen that two target systems could be placed into the same shielding block for economic reasons.

2.2.7 Production of a 5-MW Proton Beam

To increase the power of a proton beam from 1 MW to 5 MW, either the beam current or the kinetic energy (or the product of the two) must be raised by a factor of 5. The accelerator techniques required to achieve a 5-MW beam are no different in principle from those required for 1 MW. A first-stage linac followed by a compressor ring or an RCS would remain viable options, although the use of multiple rings may be advantageous. For instance, the use of three compressor rings, rather than one, has been proposed for consideration both for the 5-MW European Spallation Source (ESS) and for the 5-MW version of LANSCE-II at LANL.

In the latter case, a brief summary of the upgrades required from the 1-MW version will show that they are straightforward and require only existing technology. The general aim is to increase the average beam current by a factor of 5 from 1.25 mA to 6.25 mA, while leaving the energy unchanged at 800 MeV. Major improvements are required in four areas:

- **H⁻ ion source.** To obtain extra current, two H⁻ sources will be used, each rated at 65 mA. (The present state-of-the-art source provides ~60 mA, so some R&D will be needed.) The 400-MHz RFQ and drift-tube linac will also be

duplicated, the two beams being merged at 20 MeV to fill every 800-MHz bucket of the remaining linac tanks.

- **Linac RF Beam Loading.** The existing rf power modules can cope with some increase in current, but for a factor of 5 it will be necessary to double their number.
- **Compressor Ring Tune Shift.** The transverse tune shift due to space charge limits the number of protons which can be injected into the compressor ring, for a given energy and beam emittance. To accommodate 5 times more protons, the beam emittance is increased by 5/3, taking advantage of a 30% conservative aperture provided in the 1-MW storage ring, and 2 identical rings are added in the same tunnel. [A similar scheme has been in successful operation at CERN for many years, where the 1-GeV PS Booster Synchrotron actually consists of 4 separate rings in a vertical array.]
- **Beam Loss Control.** Fractional beam loss in the linac must be controlled 5 times more strictly, but loss control is so effective here that this should present no problem. In the compressor rings, experience gained at 1 MW should enable the losses per ring to be no higher than before. Careful attention will have to be paid to the beam lines feeding the neutron targets.

These factors have been described at some length not because this scheme is necessarily the optimum one, but because they demonstrate the technical feasibility of producing a 5-MW proton beam using presently available, even familiar technology. They also allow the cost ballpark to be estimated ($\leq \$0.5B$) to go from a 1-MW to a 5-MW PSNS.

A more accurate estimate must await Conceptual Design Studies and Reports, which could examine different design scenarios in some detail and select an optimum. In this regard, it would be helpful, though not essential, to know the ultimate beam power required of the facility. If the eventual aim were 5 MW, it would of course be more economic to design for this from the beginning, than to have to modify a 1-MW design for upgrading to higher power. Alternative designs could be considered with the linac extended to a high enough energy (~ 1.6 GeV) that a single compressor ring would suffice for a 5-MW beam.

It should be noted that 5 MW does not represent any magic barrier for accelerated proton beams. Nor are radio-frequency linacs the only candidate accelerators. At powers of 5 MW and above, induction linacs should be seriously investigated. With such machines, the beam pulses are short enough that a compressor ring is no longer needed. One such design considered for tritium production would produce 0.1- μ s pulses at 360 Hz, with an average current of 860 mA at 700 MeV — an average beam power of 600 MW, for an estimated cost of \$850M. Scaling down to a more suitable repetition rate for a spallation source, ~ 36 Hz, one would have a 60-MW beam for somewhat less cost.

In summary, it is possible to produce a 5-MW beam within the existing technology base, at a cost no more than double that of producing a 1-MW beam.

2.2.8 Target System for a 5-MW Pulsed Spallation Neutron Source

A 5-MW pulsed spallation neutron source is feasible; however, some work must be done to validate the concept.

In contrast to the upgrade of the accelerator, going from 1 to 5 MW in the target system is not simply more of the same. Since the need to retain a high source brightness does not allow one simply to increase the target dimensions, the load on the target will inevitably increase (i.e., its power density and radiation damage rate will go up) resulting in a shorter lifetime.

The most difficult problems for a 5-MW pulsed spallation source lie in the areas of heat removal from the target and radiation damage to materials. The European Community is proposing a study of a 5-MW PSNS for Europe, the ESS. Also, the LANL states that its proposed 1-MW PSNS would be upgradable to 5 MW. In the German SNQ study, a 10-MW rotating target had been worked out in considerable detail.

A recent (February 1992) workshop was held at the PSI in Switzerland to look at all technical aspects of targets and moderators for a 5-MW PSNS. The conclusion for the target station was that:

"Of the target options considered (for 5-MW beam power) the stationary water-cooled target was considered to offer the best overall properties. The design will be a technical challenge, but the working group felt that there was every prospect for success. A rotating target based on the SNQ design was recognized as a viable option which could be adopted for the new source if the difficulties with the stationary target proved insurmountable."

A 5-MW PSNS would employ two target stations, of 4- and 1-MW respectively, for reasons of duty cycle required for different types of instruments.

The specific items identified at the PSI workshop as requiring further study were divided into three categories:

- Simulation Calculations (Six man-years effort estimated)
 - Explore proton energy requirements using simple target system geometries;

- Investigate different moderator geometries including pulse shaping absorbers (poisons/decouplers/liners);
- Investigate reflector materials and performance;
- Optimize target, moderator, and reflector configuration using realistic geometry for the chosen proton beam energy to arrive at highest neutron beam intensities;
- Evaluate atomic displacement rates for the assessment of radiation damage to components; and
- Assess power deposited in all components.
- Engineering Studies (Seven man-years effort estimated)
 - Overall mechanical design of the two target, moderator, and reflector assemblies;
 - Thermal hydraulic and stress analysis for both time-averaged and pulsed conditions;
 - Cooling system design;
 - Outline safety assessment;
 - Assessment of instrumentation and remote handling;
 - Overall layout of the two target stations.
- Materials Investigations (Two man-years effort estimated)
 - Assessment of radiation damage;
 - Assessment of corrosion effects in the cooling system;
 - Assessment of radiolysis effects in cooling water and liquid methane;
 - Investigation of materials for neutron absorbers.

A total of 15 man-years would be required for all three studies.

We note that there is an ongoing effort (20 man-years) at LANL to design a target system for APT. Tungsten is used in the APT target design. A PSNS design will benefit from these APT studies. Included in the LANL effort are experiments in the following areas:

- Thermal hydraulics
 - Critical heat-flux tests in tungsten to 1.7 MW/L; and
 - Effect of a focused proton beam.
- Radionuclide production in tungsten (source term for accident analysis)
- Radiation damage
 - Tungsten and tungsten-alloy irradiations are being done with 800-MeV protons (embrittlement studies); and
 - Post-irradiation studies of structural materials (e.g., aluminum and inconel).

We also note that 5 MW is *not* an upper limit for a PSNS target system. To go beyond the 5-MW level, spallation source targets could be of the rotating solid type (SNQ) or the liquid type (Advanced SINQ and APT target designs). What is exciting about the overall accelerator spallation neutron research approach is that we have not identified the upper limit for their performance.

2.2.9 User Needs in a Neutron Scattering Facility Based on a Pulsed Spallation Source

The bulk of this report deals with technical matters related to the source. A multitude of needs must be supplied in order that an accelerator-based PSNS serves its users effectively, which this working group has addressed in a preliminary way. It is necessary to record at the outset that neutron scattering experiments, as they are typically pursued at a central user facility, require a few hours, a few days, or at the most a week or so. Active participants are either individuals or small groups. The measurements typically involve a sequence of calibration and background runs wherein the sample conditions are varied systematically (and frequently irreversibly). The experiment is unsuccessful if the series is not completed. In some cases the samples are inherently short-lived so that the experiment must be executed at a time that can be accurately anticipated.

The user facility needs fall into several categories.

2.2.9.1 Facility

The facility, the instruments, and ancillary equipment must have regular availability, more or less year-round without big gaps, to enable pursuit of rational scientific programs. When an experiment is scheduled, these must operate reliably to enable completion of the experiment. Reliability is defined as the ratio of the amount of time (in units of the time to complete one run) that the source actually operates, to the scheduled amount of time. Users are happy if this figure is greater than 95%, grudgingly satisfied if the figure is 80%, vexed and disappointed if the figure is 70%. Reliability of the source should exceed 90%.

The facility must be supported as an independent entity, with no-cost access to general users (see below regarding proprietary use).

2.2.9.2 Instruments

The facility must provide instruments enabling a wide variety of experiments at the state-of-the-art. These must be maintained and calibrated to high standards and transparent enough to be operable by general users. A wide variety of ancillary equipment must be available. Instruments and ancillary equipment must have the same high reliability as the source.

2.2.9.3 Computers

Data acquisition and experiment control computers must be reliable and friendly. Computers for data analysis must be adequate in power and number and equipped with appropriate software to enable rapid reduction of the data. Computers for *a priori* calculations for interpretation of experiments must be available and of sufficient power. The computing system should feature exportable software and off-site access to the system.

2.2.9.4 Support Services and Personnel

Users must have access to capable technicians for assistance in carrying out all (except in the most specialized) aspects of experiments. Specialist scientists at the research level and degreed scientific support personnel must be available to assist in all phases of experiments. The host laboratory must contain a broad and relevant research base to provide scientific consultation and enable research with collaborators. Office services and administrative support must be provided for visiting users.

Safety services and training mechanisms must be provided in all the required areas, which represent minimal distractions.

The program of use should be governed by an advisory committee consisting of expert members representing the entire range of applications of the facility and having a chair and a majority of members independent of the host institution. The program advisory committee reviews submitted proposals and recommends to the facility management the instrument time to be allocated. Arrangements should be standardized which allow for proprietary use (publication of results not required) at the user's expense. The facility management should be empowered to arrange specially negotiated uses, and its own initiatives.

Users must have ready access to facilities for lodging and meals. Office, sample preparation, and laboratory space for users must be provided close to the facility. The facility should be in close proximity to the majority of its users and reasonably accessible by road and public transportation.

Mechanisms should be available to provide some financial support for faculty, student, and other visitors and special support for worthy, impoverished users.

2.2.10 Summary, Conclusions, and Recommendations

2.2.10.1 Summary

The technology of pulsed spallation sources is constantly developing. The technology base for a 1-MW facility is available, with a 5-MW source requiring some R&D. The spallation working group heard presentations from LANL, ANL, BNL, and from Grahame Rees, who presented the outlook on spallation sources in Europe, primarily ISIS and the ESS. They were also presented with an update on the status of targets by working group members with considerable expertise in the field. Two laboratories presented costs for a 1-MW source using their existing assets and facilities. Consideration of all this information led the working group to conclude that for a total estimated cost (TEC) of \$0.5B, a 1-MW source could be built.

2.2.10.2 Conclusions

1. Spallation sources have been remarkably successful (ISIS at 120 kW, 800 MeV; LANSCE at 62 kW, 800 MeV; and IPNS at 7 kW, 450 MeV).
2. There is a rapidly growing user community attracted by the expanding scientific opportunities.

3. A 1-MW source consisting of a ~ 0.4 -GeV linac + synchrotron *or* ~ 1 -GeV linac + compressor ring plus associated target and beam lines can be built using the currently demonstrated technology base. Utilizing existing assets/infrastructure, this can be done at an additional cost (TEC) of $\sim \$0.5$ B.
4. A 5-MW source consisting of a ~ 1 -GeV linac + 3 compressor rings, or a higher energy linac with single storage ring is feasible. The targets for such a facility will require some R&D to address cooling and radiation damage issues.

A careful conceptual design study is expected to show that the upgrade of a 1-MW facility to a 5-MW facility can be built for an additional $\$0.5$ B if suitable provisions are made in the beginning.

5. Alternative approaches such as using an induction linac or superconducting cavities should be evaluated.
6. Steady-state operation including user support requires $\$50$ – 60 M per year for a 40-week scenario.

2.2.10.3 Recommendations

Need for Conceptual Design

1. A 1-MW pulsed spallation neutron source is desirable and appears feasible with existing technology, based upon the preconceptual designs presented by LANL and ANL, as well as information gathered from the ESS study, and target information provided by working group members. Careful Conceptual Design Reports urgently need to be prepared to better define the cost, scope, and schedule for such a facility.
2. Close interaction with the world neutron community is essential.

3 Neutron Scattering Applications

3.1 Condensed Matter Physics

G. Aeppli (AT&T Bell Laboratories)
R. Cowley (Oxford University, U.K.), Chairperson
J. Lynn (University of Maryland)
P. Sokol (Pennsylvania State University)
H. Zabel (University of Bochum, Germany)

The existing DOE neutron scattering facilities do not meet the needs of condensed matter physics in the U.S. This situation can only be remedied by the construction of a new high-flux reactor and a state-of-the-art spallation source, which would allow a full complement of neutron instrumentation. However, if the construction of only a single source is possible, then the choice depends on the time-averaged flux which can be actually realized at each facility. If the reactor-to-spallation-source flux falls within the guidelines of 50:1 or better, then a steady-state source is clearly optimum for most experiments, while if the ratio drops to 5:1 or smaller, then a spallation source would provide the best overall capabilities. In the short term, we recommend that the instrumentation at existing facilities be upgraded and the facilities run more reliably and with better accessibility to the neutron scattering community. We also recommend an aggressive program of instrument development be undertaken.

3.1.1 Scientific Case for Neutron Sources

The importance and uniqueness of neutrons for studies in condensed matter physics has been well documented in many reviews and assessments, and therefore here we intend simply to update these assessments. The working group considered that this importance has clearly been strengthened over the last few years, and is confident that neutron scattering will continue to be an essential probe of condensed matter systems for the foreseeable future. In reviewing recent experiments, we felt it was misleading to select a few individual experiments to represent the entire field. Rather, the nature of neutron scattering is such that it enables not only pioneering experiments to be performed, but that it also has a broad impact on many important areas of condensed matter physics.

One of the most important uses of neutrons is to develop the understanding and technology of new materials as they are discovered. When fundamentally new materials, such as the high-temperature superconductors or the fullerenes, are discovered, neutron scattering is the choice of probes to investigate the physical properties. These range from the basic atomic and magnetic structures to the dynamics of the atoms and the magnetic moments. This is well-illustrated by key neutron studies of the high-temperature superconductors which determined the atomic properties (in particular the oxygen positions and concentration which are central to understanding the superconductivity) as well as the role of phase stability, the formulation of impurity phases, the nature of the magnetism, and the lattice dynamics. Other examples of new materials for which

neutron scattering has provided crucial information are: heavy-fermion systems, where neutrons revealed unique information on the magnetic properties; quasicrystals, where the atomic and magnetic structure, and vibrational frequencies have been determined; and the fullerenes, where the structure, the lattice and rotational dynamics, and the phase transitions have been determined.

While the information obtained in neutron studies is vital to the understanding of new man-made materials, neutron scattering also provides a steady stream of new discoveries in familiar materials. Recent examples are the characterization of the structural consequences of bone mineralization, where the diffraction studies have provided unique insight into the role of calcification in bone structure and strength that was not obtainable with any other technique. Another example is the structure and dynamics of hydrogen in metals, including the quantum mechanical tunneling of the protons between double-well potentials, wherein neutron studies played a central role in the physical analysis of these technologically important materials.

Most materials, particularly those of technological interest, are not single crystals; they are either polycrystalline or have a randomized or disordered structure. Neutron scattering has provided unique information about the subtle and unexpected properties of such systems. Crucial experiments have been performed on a variety of glasses, providing information on the structure, domain sizes, and dynamics of spin glasses and disordered ferromagnets. Similar experiments on dipolar glasses have shown that the molecular dipoles can become frozen in either ferroelectric, random, or antiferroelectric arrangements. Studies of conventional glasses, which are amorphous materials, have shown how the phonons differ from those in crystalline materials. The effect of random fields on the ordering of magnetic systems has led to the discovery of an unexpected region of metastability below a well-defined temperature.

Neutron scattering, while vital to the understanding and development of new materials, also plays an important role in elucidating the fundamental properties of condensed matter systems. These fundamental problems are often concerned with the solution of simply posed problems in quantum mechanics and statistical mechanics. Arguably one of the most fundamental systems in condensed matter physics concerns the liquid and solid phases of ^3He and ^4He . Experiments with neutrons have provided uniquely important information on, for example, the Fermi liquid parameters of ^3He , the Bose condensation of liquid ^4He , and the magnetic structure of solid ^3He . Another fundamental area in which there has been recent progress is low-dimensional systems, where both one- and two-dimensional systems have been actively studied. Neutron scattering has shown that one-dimensional spin systems are excellent realizations of spin liquids, with qualitatively different properties depending on whether the spin is integer or half-integer. In particular, the excitation spectrum has a gap, the Haldane gap, for $S = 1$, while for $S = 1/2$, the spectrum is dominated by pairs of spinons rather than linear spin waves. Experiments on the high- T_c superconductors have led to a detailed understanding of the behavior of two-dimensional (2-D) Heisenberg systems and to the effect of the metal-insulator transition on the magnetism.

One of the increasingly important areas in condensed matter physics is that of layered and multilayered structures, whether produced naturally or by means of Molecular Beam Epitaxy or Metal-Organic Vapor Phase Epitaxy techniques. As an example, artificially structured layered

materials are essential to important new device applications, such as magnetic recording media, or devices based on the giant magnetoresistance property such as recording heads. Neutron scattering has provided detailed information about the magnetic structure on the nanometer scale, which has shown, for example, that the magnetic order can propagate through nonmagnetic layers, and that the giant magnetoresistance is not necessarily associated with antiferromagnetic couplings.

Finally, an important area has been the development of neutron scattering techniques for the study of surfaces and buried interfaces. In the area of condensed matter physics, this provides great possibilities for the study of surface effects in magnetism, superconductivity, and liquids. These types of experiments have already been very productive, and promise important advances, particularly if the techniques and fluxes are improved.

3.1.2 Comparison of Spallation and Reactor Sources

The best choice for a source depends on the number of useful neutrons that strike the sample. The limiting cases for comparing a spallation source with a reactor are clear; if the reactor flux exceeds the peak flux of the spallation source then it will clearly be superior, while if the time-averaged flux of the spallation source exceeds the reactor flux then the spallation source is to be preferred. We have assumed, for the purpose of comparing potential future sources, the same parameters set forth in the Shelter Island study.¹ Thus, we will compare the relative advantages and disadvantages of a reactor source with a spallation source having 10 times the peak flux and 1/50 of the time-averaged flux. Our conclusions depend on these assumed parameters and will change if the ratio of time-averaged fluxes changes as discussed below.

The experiments of interest in condensed matter physics span a broad range of momentum and energy transfer, and impose a variety of, often conflicting, requirements on the instrumentation required. The necessary capabilities can be broadly characterized by the requirements for wave vector Q and energy transfer and the ability to resolve these quantities while still maintaining sufficient signal-to-noise. Also of central importance is the type of sample (i.e., single crystal, powder, liquid, etc.) and whether polarization analysis is desirable. Below we characterize in broad terms the instrumentation needed for different types of measurements and discuss the relative merits of reactor versus spallation sources for the measurements.

1. A wide class of measurements in condensed matter physics requires the measurement of scattering at a particular point in Q - ω space, or along a well-defined path in this space for single-crystal samples. In general such measurements are limited to neutron energies below a few hundred meV. Examples of such measurements include the determination of the order parameter in an antiferromagnet, or the study of particular phonons or magnons. These experiments have been traditionally carried out using triple-axis spectrometers and, in all likelihood, this will continue to be the instrument of choice on any future source. The success of triple-axis type experiments is most directly related to the total time-averaged flux available. On this basis, the reactor source is superior because of its larger average flux. However, we note that spallation sources can eliminate some of the difficulties associated with reactor-based measurements, such as $\lambda/2$ contamination.

Thus based on these considerations, these measurements are best carried out on a reactor source unless the total time-averaged flux of the spallation source is close to that of the reactor.

We also note that, in some circumstances, spallation sources do provide special advantages for these types of measurements. A recent example of this type of measurement is the determination of the phonon dispersion relations in La_2CuO_4 . In the past these measurements have been performed at reactors with triple-axis machines. Nevertheless, we consider that time-of-flight (TOF) techniques should be an effective way of performing these measurements in the future, but this will require the development of large-area, high-resolution position-sensitive detectors (PSD's) as well as the development of far more sophisticated computational techniques for analyzing and displaying the data effectively.

2. Another broad class of measurements are those that require large momentum and/or energy transfers. Examples of these types of measurements are studies of high-energy magnetic or lattice-dynamical energies, momentum distribution studies, and measurements of the structure factors in amorphous systems. In view of the high incident neutron energies required for these measurements, spallation sources are preferred because of the copious production of epithermal neutrons. A reactor with a hot source can enhance this region of the source spectrum, but even a small spallation source would have superior performance for most of these measurements. Thus, these measurements are clearly best carried out with a spallation source.

3. Another broad class of measurements are those where a large region of Q - ω space is scanned without regard to the direction of Q . Traditionally, this is where TOF techniques with a wide range of detector angles are particularly well suited. Examples include the excitations in liquids and amorphous solids, or phonon density of states measurements on polycrystalline samples. These TOF techniques are well established at both reactor and spallation sources, but with the assumed 10:1 peak flux advantage of the spallation source, it will be the clear winner for these types of measurements. However, in some cases reactors may be more desirable, for example, if the experiment can use a faster repetition rate or a longer time pulse. These can in principle be obtained with a reactor source but not with the spallation source, so that the gain in using the higher peak flux from the spallation source will be correspondingly reduced.

Our overall view is that a spallation source is superior for these types of measurements if the peak-flux ratio is 10:1 or higher, while if the ratio is between one and ten, then either source may be used effectively to carry out these measurements.

4. A broad class of measurements are those where we require excellent Q -resolution but are relatively unconcerned with energy resolution. In general, these are diffraction measurements such as on single crystals or powders, small angle scattering, and reflectometry. Examples of successful instruments for each of these techniques exist on current reactor and spallation sources. Diffraction instruments at reactor sources generally make use of crystal monochromators in the incident beam and either scan the detectors or make use of multidetector arrays or PSD's. Thus, these instruments would benefit most from a higher time-averaged flux. Alternatively, diffraction

instruments at spallation sources are of course TOF machines, with detectors covering a wide angle range to increase the data rate. These instruments are well suited to spallation sources.

For sources with a time-averaged-flux ratio between 50:1 and 5:1, similar quality of results can be obtained. At present the techniques are somewhat better developed at reactor sources, particularly for single-crystal structures and small-angle scattering. Nevertheless, we expect that continued development at spallation sources will close this gap in the future.

Two further comments are in order. Firstly, we have based our comparisons on the assumption that future spallation sources will have a broad range of moderators available that provide neutron beams with different spectral distributions. In particular, we have assumed that cold moderators will be available. While we foresee no problems in principle, this technology has not been extensively demonstrated on spallation sources at this time. We note that the trend in condensed matter science is towards investigating more complicated systems, which are then larger and heavier. Hence it is likely that the historically increasing demand for cold neutrons will continue in the future.

Secondly, it is clear that the use of polarized neutrons will play an increasing role in the future, and thus the ability to use polarized neutrons is an important consideration. These techniques are relatively well developed at reactors at present, especially at long wavelengths, and technological advances can be expected to make them more efficient. However, there is at present no effective way of producing broadband (white-beam) polarizers which are necessary at spallation sources, except at long wavelengths. While research is underway on broadband polarizers, the availability of these devices in the future is speculative, and hence this currently represents a disadvantage for pulsed sources.

In summary, the proposed reactor and spallation sources are complementary in that many of these experiments will be more effective on one or the other. For example, triple-axis studies of single crystals, where a particular path in Q - ω space is desired, are generally best performed at a reactor. Alternatively, measurements requiring high energy or momentum transfer, such as high-energy magnetic excitations or structure studies of amorphous systems are best performed at a spallation source. These conclusions do not depend strongly on the assumed flux ratio. For example, only if the average-flux ratio were smaller than 5:1 would a spallation source be comparable to a reactor source for these types of measurements.

These two types of sources also overlap strongly in a number of areas. For example, single-crystal and powder diffraction, small-angle scattering and quasielastic scattering can be effectively carried out at either source. For these measurements, the advantages and disadvantages of either type of source is less clear. The crossover occurs in the range $5 \rightarrow 50:1$; the exact crossover depends on the particular instruments and measurements required and must be determined on a case-by-case basis. Thus, in this region there is no overwhelming advantage to either source.

We conclude that each type of source offers considerable advantages for some types of measurements, while others can be performed equally well on either type of source. The community clearly needs both types of source available. If it is necessary to choose one source, the view of the working group is that, if the time-averaged flux of the reactor to spallation source is 50:1 or larger, then a reactor source is superior for most studies in condensed matter physics, while if the ratio is 5:1 or less, then a spallation source is to be preferred. In between these ratios, the choice depends on the balance of interests and the perception of development potential, and so it is difficult to choose between the sources objectively.

3.1.3 New Opportunities with a High-Flux Source

Neutron scattering is a unique and exceptionally useful probe of condensed matter, and it is abundantly clear that this will continue for the foreseeable future. Moreover, the development of other techniques which provide complementary information, such as x-ray scattering, will result in even more, complementary, neutron experiments being required, rather than less.

A tenfold increase in available neutron flux will make many new experiments possible. The most obvious are those that become possible with small samples. It is important to realize that the current sample size requirements for inelastic scattering have generally delayed and on some occasions prevented neutron studies of many important phenomena in condensed matter physics. Broadly construed, small samples also include:

1. Surfaces and interfaces of systems ranging from liquids to magnets to superconductors.
2. Weak scatterers such as carriers at low density in semiconductors and dilute impurities.
3. Strong absorbers.
4. Samples in extreme environments, such as high-pressure cells, which can be both absorbing and/or of small volume.

A second category of new experiments, which will take advantage of higher source flux, are those that yield more information about larger samples. These include:

1. High-Q- and E-resolution experiments. Neutron scattering is unique in its ability, frequently with the same instrument, to obtain a large range of resolution profiles. More flux will increase this range. High-Q-resolution x-ray scattering has actually increased the demand for medium- and high-Q-resolution neutron experiments.

2. Polarized beam experiments, which have so far been focused primarily on magnets, have great promise in separating incoherent and coherent scattering from organic materials.
3. Kinetic studies with high-real-space resolution.
4. Microscopy and tomography, taking advantage of both magnetic and nuclear couplings between neutrons and materials.

The above examples are generally extrapolations of what is done with neutrons today. We also look forward to moderator, beam transport, and instrument advances which together with the new sources will produce effective flux improvements of *several* orders-of-magnitude in portions of the source spectrum, and certainly these types of measurements will benefit not only quantitatively but qualitatively from these enhancements. Far more exciting are entirely new fields that would be opened with new high-intensity sources, such as ultracold neutron (UCN) spectroscopy, neutron microscopy, studies of nonequilibrium phenomena, and investigations of excitations at high energies. We are convinced that these fields, representing the hot and cold frontiers of neutron scattering, will bring excitement and crucial knowledge about condensed matter physics for a long time to come.

3.1.4 Existing Facilities

There are serious problems with the availability of the DOE sources both currently and in the recent past, as well as the nature of the instrumentation and the accessibility of the current DOE facilities for neutron scattering research. This is a distinct disadvantage to U.S. scientists. Improvements need to be made to accommodate the neutron scattering community until new neutron sources can be brought on-line.

3.1.4.1 Availability of the Sources

In recent years the BNL and ORNL reactors have been shut down for considerable periods, and the LANL facility has only operated for a few weeks each year. It is clearly not cost-effective, considering the large capital investment in sources, instrumentation, and personnel, for these facilities not to be operated. It is also extremely difficult for these facilities to develop and maintain a viable user community under these circumstances. Resources should be made available so that the existing facilities can be run in an efficient manner.

3.1.4.2 User Access

It is clear that the range of problems explored with neutrons has expanded in European countries as a direct result of their strong user base. A larger user community in the U.S. will make better use of the facilities and enable more U.S. groups to take advantage of the unique information provided by neutrons, particularly in areas where a multidisciplinary approach is needed, such as the development of new materials. This requires that access to the DOE facilities be made easier, that the facilities run on a well-defined schedule allowing users to arrange their home commitments around the available beam time, and that financial support be made available to cover the travel and living expenses of users.

3.1.4.3 Instrumentation

The instrumentation at the present DOE neutron facilities is lacking in two respects. Firstly, the instrumentation often does not incorporate new developments that would provide higher flux on the sample and greater experimental flexibility, which would be equivalent in some cases to an effective flux increase of an order-of-magnitude. An example is to have available a selection of remotely selectable, variable-focusing monochromators on triple-axis instruments. Secondly, the existing instrumentation is limited in scope since new instrument development has not been aggressively pursued, primarily due to a lack of resources. Both these difficulties restrict the range of studies that U.S. researchers can carry out with respect to their European colleagues; one recent illustration is provided by the determination of phonon frequencies in a 3-mm³ fullerene crystal that was carried out at a medium-flux reactor in Europe.

We recommend that a substantial effort be made to modernize existing instrumentation. This modernization will help provide for the needs of the community until new sources are available. These improvements should include new beam optics, more detectors on TOF instruments, and increased computational facilities.

We also recommend that a larger variety of instruments be made available. For instance, there are no spin-echo or backscattering spectrometers available in the U.S., limiting the studies that can be carried out by the physics, chemistry, biology, and materials science community. Although new instruments at the National Institute of Standards and Technology (NIST) will, in the near future, address some of these inadequacies, the expected demand will quickly exceed the available time.

The development program for instrumentation should not be limited solely to building new instruments. Programs should also be initiated to develop better monochromators, high-speed choppers, beam-focusing and polarization technology, and new detector systems. These systems will certainly be required for the next-generation source, and can provide immediate benefits on existing sources. We note that these modernization and development programs will not be successful without an increase in staff at the DOE facilities.

Finally, we note that much of the cutting edge science is governed by the ancillary equipment available; the ability to provide extremes in temperature, pressure, and magnetic fields is essential. This equipment is often expensive and complex and must work reliably if expensive beam time is not to be wasted. We recommend that sample environments facilities be substantially improved and that adequate support for these facilities be provided. These improvements are essential if continued cutting-edge research is to be carried out at DOE facilities.

If the modernization of the existing DOE facilities is not implemented to improve the facilities for U.S. neutron scattering until a new source is constructed, we recommend serious consideration be given to buying time at the ILL and ISIS to provide access for U.S. scientists to the special facilities available there while keeping the existing U.S. facilities in operation.

3.1.5 Reference

1. G.H. Lander and V.J. Emery, *Nucl. Instrum. Methods B* **12** (1985), 525-561.

3.2 Materials Science

T. Holden (CRNL, Canada)

G. Long (NIST)

S.C. Moss (University of Houston), Chairman

J. Parise (State University of New York at Stony Brook)

J.R. Weertman (Northwestern University), Panel Member

B.J. Wuensch (Massachusetts Institute of Technology)

We consider it important at the outset of this brief report to define the scope of our inquiry into the developments in neutron scattering as they apply to Materials Research. In one sense all matter explored by scattering falls within the category of "materials". We have, however, limited ourselves, through discussion with the Panel, to selected areas on which we feel competent to comment and which we believe do not overlap too much with the other working groups. Many of these areas will also have a strong bearing on technologies which they support, either directly or indirectly, and we will try to illustrate these connections in the text. In this sense, then, we have construed "materials research" as that which may ultimately relate to technology or shed light on a field which is of general technical interest.

We would also like to emphasize that the neutron scattering we are all discussing here is small science done at an expensive source whose cost must be assessed against the large number of scientific programs and users that it serves. In other words, a group of researchers will visit a scattering center from a few days to, perhaps, two weeks on a single experiment and then return home to reduce and analyze the data. This mode of operation differs significantly from, say high-energy or particle physics.

To start with, we shall extract from the Shelter Island summary¹ selected features relating to materials research and discuss very briefly the progress made on them to date. We will then highlight several important achievements in the past decade, address the relative strengths and weaknesses of the current sources for materials research, and project new opportunities for a " $\times 10$ -flux" source. Finally, we present a list of shorter term requirements for improved instrumentation.

3.2.1 Update of the Shelter Island Report

Section 3.2 Industrial Applications of Neutron Scattering

There has been considerable progress made here in particular on:

- a. structure and conformation of polymers (discussed elsewhere).
- b. hydrocarbon fluids in porous media and gases on surfaces; intercalates and related systems.
- c. growth of precipitates and voids; cracks and defects in metals and ceramics (although real-time studies remain for the future high-intensity sources).
- d. The use of H_2O/D_2O contrast to understand crystallizing aluminosilicate gels in the presence of tertiary amines (templates); this process attempts to tailor-make cavities in these framework materials.
- e. The use of quasielastic scattering to study diffusion of fluids in restricted geometries.
- f. The use of small-angle neutron scattering (SANS) to study phase separation in materials is now quite well developed but, again, time-dependent studies require, except in isolated cases, much greater flux (see bottom of *Section 4.1*). Nonetheless, there have been excellent studies of the time evolution of phase separation in alloy systems (Mn-Cu, Fe-Cr) using both spallation and steady-state sources.

Section 4.1 Resolution

- a. In SANS experiments, we still do not have a Q-resolution of $\sim 10^{-4}$; in other words, the relevant ranges to be discussed, and which are currently available with synchrotron x-rays, are not yet achieved.
- b. Residual stress measurements now represent a technique used routinely at both spallation and reactor-based sources on samples of 5-50 mm³.

Section 4.2 Small Cross Sections and Differential Scattering

- a. The study of materials at high pressure has been accomplished with a Besson cell to a maximum of 150 kbar at room temperature; studies at simultaneous high pressure and elevated temperature await development.

Section 4.3 Polarization Analysis

- a. Using supermirrors developed since 1984, polarization analysis of neutron reflectometry at NIST has been done, for example, to understand the passive (oxide) layer on Fe under potentiostatic control.

Section 7 Small-Angle Scattering

- a. SANS has already been referred to, and it has been noted that the Q-range is still limited, especially at the pulsed sources. It would be "desirable" to achieve $Q \leq 1 \times 10^{-4} \text{ \AA}^{-1}$ routinely. Nonetheless, SANS is still the technique most in demand at all sources.
- b. Densification of ceramics as a function of sintering has already been studied but *in situ* studies of aging kinetics, of precipitate growth under thermal processing, and the response of ferrofluidic systems to applied magnetic fields await more intense sources.

Section 8 Collective Excitations

In particular, we note here that the scattering law, $S(Q, \omega)$, for superionic materials, hydrogen in metals, plastic crystals and orientational glasses (class C experiments) has been studied in some detail. The quasielastic response of many of these materials is now known and the glass transition in more traditional glass-forming liquids has also been explored. Again flux is an issue if good Q- and ω -resolution are simultaneously required. In addition, inelastic scattering has been used to great effect in studying precursor phenomena above selected first-order structural phase transformations.

Purely elastic scattering has permitted direct evaluation of short-range order and static displacements without the complication of the phonon contribution to the diffuse scattering.

Finally, crystallography at a pulsed source permits all of reciprocal space to be explored, including diffuse scattering where it is of interest. This is of particular importance when the important regions of reciprocal space have yet to be identified. The method is flux-limited until larger crystals become available.

Section 9 Crystallography (powder)

Current estimates of the maximum number of fittable parameters in a Rietveld analysis are ~150.

Time-resolved studies can be done on a time scale of minutes; again, more flux is still required for seconds.

Section 11 Liquids and Amorphous Solids

Current work includes D4 at ILL where, for substantial sample volumes, isotopic substitutions have been accomplished for the separate constituents of several binary liquids and glasses. The study of quasicrystal structure is currently being advanced significantly through double isotopic substitution in Al-Cu-Fe powders to permit the collection of several hundred peaks each from six separate samples of the same composition at ISIS with preliminary work at NIST.

The structure and dynamics of glasses remains an important topic, but the latter is still currently flux-limited. The structural aspects, however, have been substantially enhanced through higher Q data at ISIS and IPNS.

In summary, the Shelter Island report remains largely an accurate statement of the conditions in neutron scattering vis-à-vis materials research. We note that excellent progress has nonetheless been made in selected important research areas, some of which were naturally unknown at the time. However, because the infrastructure was in place, it permitted an immediate response to the topical issues; we should not lose sight of this feature in future planning.

3.2.2 Achievements of the Past Decade

3.2.2.1 Neutron Powder Studies of High- T_c Superconductors

Since the fall of 1986, high-temperature superconductors have been a dominant feature of the scientific and technical scene. Essential to an understanding of these perovskite-based materials

has been a constant output of (largely powder) neutron diffraction studies detailing their structure. Neutrons have been particularly important in determining the oxygen concentrations, positions, and thermal ellipsoids in the presence of heavier atoms (La, Ba, Y, Sr, Cu, Tl, Bi, Pb, etc.). The principal technique has been Rietveld refinement of high-resolution powder diffraction data, mainly at pulsed spallation sources (IPNS, LANSCE, ISIS) but also at steady-state sources such as at NIST and the ILL. It is clear that the precision/accuracy of these structure refinements, particularly as regards the oxygen, would have been considerably inferior using powder x-ray data. As examples among the many issues that were of considerable concern are:

- a. The degree and importance to superconductivity of oxygen stoichiometry and ordering in the chain sites of the CuO plane of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and phases related by doping and substitution.
- b. The relationships of structure and stoichiometry to the magnetic properties of these materials (i.e., in $\text{La}_2\text{CuO}_{4+x}$, x was essential in establishing the superconductivity and in deciding the balance between high T_c and antiferromagnetism). In $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, similar issues obtain in the oxygen-depleted insulating phase.
- c. Oxygen ordering also seems recently to have been revived, through powder diffraction studies, as a potent influence on T_c .
- d. The phase diagram and crystal structures of the $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ system were determined and were related to the semiconducting-superconducting transition in this material at $x \simeq 0.37$.
- e. Recently a new generation of superconductors has been discovered whose T_c is above 40 K. The materials, $(\text{Ca},\text{Sr})\text{CuO}_2$, synthesized under pressure and above 1000°C in excess O_2 are available in small quantities at ambient temperature and pressure and are currently beginning to be studied by neutron powder methods.
- f. Very recently at ISIS, the high-frequency portion of the dynamic structure factor, $S(Q,\omega)$, in powders of $(\text{La},\text{Sr})\text{Cu}_2\text{O}_4$ was integrated and transformed to give an oxygen-weighted pair-distribution function, $g(r)$, which showed a prominent anomaly at T_c . This is an extremely interesting use of a pulsed source.

3.2.2.2 Multiple Small-Angle Neutron Scattering During the Densification of Ceramics

Multiple small-angle neutron scattering (MSANS) has been successfully applied to the study of microstructure evolution as a function of thermal processing (sintering) of ceramics. Such studies are a critical element in the technological goal of bringing intelligent processing to ceramic science. The creation of the MSANS formalism has made it possible to characterize closed or open porosity in the size range between 0.08 and 10 μm in materials that are 55 to 99% dense and several millimeters thick. This achievement would not have been possible before the availability of copious quantities of long-wavelength neutrons from a cold neutron research facility. In alumina, for which the diffusion mechanism dominates the sintering, it was found that the initial connectivity that is established in the green body plays a dominant role in establishing the size of the open-channel diameters that persist throughout the intermediate stage of sintering. The role of MgO as a sintering aid lies, at least in part, in its ability to prolong the stability of the intermediate sintering stage such that the ceramic body achieves greater density before the isolated porosity of final-stage sintering is formed. In silica, for which the viscous flow mechanism dominates the sintering, it was discovered that there is a transition in the pore morphology from cylindrical to disklike as sintering proceeds. This transition is very significant in that it enables the persistence of open, interconnected porosity out to rather high densities (i.e., greater than 95% of theoretical density).

3.2.2.3 Light Element Contrast

A number of technologically important materials contain light elements in the presence of heavy metals. Examples include H- and Li-battery materials; fast ionic conductors such as AgI, Ag₂S, UO₂, and ZrO₂; toughened ceramics (Y₂O₃/ZrO₂); and magnetic ferrites (NiFe₂O₄ and MnFe₂O₄). This later material is the basis of a \$4B magnetic recording industry.

The neutron's sensitivity to light elements and magnetic fields, relative to x-rays, gives neutron scattering a distinct advantage in the studies of these materials.

3.2.2.4 SANS Studies on Porous Materials

SANS has proved powerful in the study of porous materials in a number of ways:

- a. Although many porous materials of technological interest (cements, clays and porous rocks, porous catalysts and ceramics, graphites, etc.) are multiphase in nature, the dominant scattering interface in the system is that between pore and solid. Thus small-angle scattering has been used to probe the pore/solid morphology over a large range of length scales. Unlike transmission electron microscopy (TEM) methods which give a highly detailed but not necessarily

representative local picture, SANS yields microstructural parameters statistically representative of significant sample volumes. Using recently developed fractal theories (which only need approximate the sample microstructure), the small-angle scattering has been interpreted to determine parameters such as the effective pore size distribution, pore surface distribution in different parts of the morphology, and relative predominance of different parts of the microstructure (e.g., intraparticle vs. interparticle porosity, etc.).

- b. By conducting real-time experiments (which require high flux, low background, and a flexible choice of instrument wavelength and geometry), it has been possible to monitor the time dependence of microstructural changes such as occur during cement hydration. Determination of the time dependence of changing surface areas or pore/particle sizes is important because kinetic theories predict different time dependencies for different mechanisms (e.g., nucleation vs. surface growth, coarsening vs. particle growth, etc.). SANS has proved particularly powerful in distinguishing the time dependencies in different parts of the microstructure.
- c. Using H/D substitution in water and methanol pore fluids, it has been possible to conduct contrast variation experiments on cements, clays, and porous rocks to determine pore-fluid access under different conditions. With a purpose-built permeation cell, the pore-fluid exchange has been followed as a function of permeation time. SANS has proved unique in providing information on pore access in different parts of the microstructure, and hence in being able to differentiate between pore-volume and pore-surface access. This ability is technologically significant because sorption processes and leaching are controlled by the accessible surface area, and most of this is in the small pores whose total volume may be insignificant. Thus, conventional sorption measurements, based on volume-averaged slurrying or through-diffusion, can give misleading results. The time dependence of the fluid exchange has allowed permeation and diffusion-driven processes to be distinguished.
- d. The internal pore structure of porous materials and the phase and transport behavior of fluids through such materials are also problems of direct interest to technological areas such as oil recovery, catalysis, separations, purification, and filtration, etc. Contrast-matched *binary* fluid mixtures can be introduced into the porous medium and the wetting of the internal surfaces by one of the components can be studied as a function of concentration or temperature by seeing the scattering from the resultant film alone. The effect of the microporosity on the transformations occurring in the imbibed fluids can be studied (e.g., phase separation in binary liquid mixtures, or near the liquid/vapor critical point, or crystallization). In all of these cases, interesting differences from such phase transformations occurring in bulk have been found by neutron scattering and other techniques. Higher intensity sources and the concomitant increased Q-resolution available in SANS experiments will enable

such studies to be extended to materials with much larger internal pore size (0.1 to 1 μm) which are relevant to industrial processes.

3.2.2.5 Residual Stress and Texture

Measurements of residual stress and texture are routinely carried out at five neutron sources in North America by both conventional and TOF diffraction. The work is generally carried out in direct response to industrial requirements and covers components as diverse as rail tracks, rocket casings, pressure tubes, weldments, and gas-turbine engine components. Current scattering volumes are of order 5 mm³ for reactors and 50 mm³ for spallation sources for roughly the same counting times and statistics. The time-averaged number of neutrons falling on a sample in the same setup is 5×10^6 n/s monochromatic and polychromatic for a reactor and spallation source, respectively. If a full complement of, say, 12 peaks is measured, this would take equal time on both sources. If only 2 or 3 peaks are needed to get a good approximation to the engineering strain field, there is an advantage to the reactor measurements.

A flux increase of $\times 10$ will give a $\times 3$ improvement in spatial resolution which will be important for measurement of near surfaces where strains vary over a scale of tenths of a millimeter, or strains for example near small rivets, or near blades on a turbine compressor. Alternatively, a threefold improvement of angular resolution will permit reducing the uncertainty in strain measurements in ceramics to $\pm 0.3 \times 10^{-4}$ where strains of technological importance are several times smaller than in metals. Noninvasive temperature measurements by neutron diffraction have shown a practical probability, with applications to the determination of temperatures within engines. Higher fluxes are required to make the method useful since times of interest, for example the time for the engine to develop full thrust, are of the order of minutes, whereas the measurements take some 30 times longer.

3.2.2.6 Martensitic and Related Structural Transformations

A major advance in the application of neutron scattering to traditional materials research is in the area of martensitic and related structural transformations in metals and insulators. Of course, neutron studies of classic structural transitions, such as SrTiO₃ and BaTiO₃, as well as the study of soft phonon modes in superconductors (Nb₃Sn) have represented striking advances in condensed matter physics. The martensitic transformations, however, are often strongly first order in character and represent shear and volume changes with often substantial barrier heights. In addition, as in the shape memory alloys (i.e., NiTi) and traditional steels, these martensitic and related transformations often have very important technological applications.

Such transformations are ubiquitous and neutrons have often proven quite useful in elucidating certain characteristic features. The basic motivation is in the search for premonitory elastic anomalies in the phonon spectra (dispersion curves) which provide insight into the

underlying physics of the transformation. We discuss here briefly one example where neutrons have proven useful, i.e., $\text{Ni}_x\text{Al}_{1-x}$ where $x \approx 0.63$.

On these alloys the transformation is from a CsCl (excess Ni on the Al sublattice) to a rhombohedral 7R structure. This transformation can be accomplished by a set of transverse shuffles in the bcc lattice of (110) planes in a $\langle 1\bar{1}0 \rangle$ direction (normal to the new c-axis). This is accompanied by a "Bain strain" which essentially converts the (110) plane(s) into close-packed planes. In the phonon spectrum, there is a pronounced dip in the classic soft bcc mode $[110]; \langle 1\bar{1}0 \rangle$ at $\sim 1/6$ of the Brillouin zone boundary or near the 7R position. On lowering the temperature, the dip lowers and elastic scattering appears at the soft wave vector, growing in intensity as the transformation is approached. Finally, the martensitic transformation takes place, but these premonitory effects give strong evidence of an anharmonic phonon mechanism, "nucleating" on defects (concentration fluctuations and static disorder) with a very soft (temperature-dependent) elastic response. In addition, recent electronic structure calculations indicate that the observed soft wave vector may originate in the electron-phonon interaction via $2k_F$ spanning vectors.

Similar effects are also seen in technologically important alloys of Zr and Ti which may undergo the well-known omega transformation. For the pure elements, no true elastic scattering is observed at the soft wave vector; on alloying with a variety of elements, the premonitory elastic diffuse intensity appears.

3.2.2.7 Characterization of Microstructural Features and Damage

We discuss here microstructural features such as precipitate and dispersoid populations, or damage in the form of Ostwald ripening of strength-giving particles, microcracking, grain boundary cavitation, or embrittlement resulting from cluster formation. The sensitivity of SANS to distributed damage indicates its value as a nondestructive method to evaluate materials integrity.

An ongoing SANS study of precipitation behavior of metal carbides in advanced high-strength, high-toughness steels is yielding valuable information on the time and temperature dependence of the nucleation and growth of these nanoprecipitates that can be used to guide future alloy development and processing schedules. Calculations of size distributions from SANS measurements, to produce data on critical nucleation size of the metal carbide, showed that their subsequent growth is in accord with models of precipitation from a supersaturated solution and revealed a surprising burst of secondary nucleation. The coherent-incoherent transition is signaled by a marked change in scattering length density contrast. (It is important to maintain coherency for maximum strengthening, as well as to resist coarsening.) The success of this study depended on the complementary use of atom probe field ion microscopy (APFIM; for the chemistry of the metal in the metal carbides, since it changes with heat treatment) and TEM (to determine the shape of the metal carbides, which also changes). While carbide sizes can be obtained from TEM and APFIM, the number of carbides sampled is 100, compared to 10^{16} by SANS.

Lower Q values are needed in the SANS instrument for the steel work in order to study the other, larger carbides. In the case of damage measurements, cavities and cracks become invisible by SANS as they grow to a dangerous size. A minimum Q of $10^{-4} \text{ \AA}^{-1}$ is therefore needed to insure that objects in the $1 \text{ }\mu\text{m}$ size range (a typical dimension of many microstructural features) can be studied.

3.2.3 Relative Merits of the Two Sources

For the topics considered in this report, both sources are useful. However:

1. SANS will be best done on a reactor cold source with tight Q -resolution.
2. High-pressure studies are currently envisioned to be best handled at a spallation source.
3. Liquids and glasses will always take inherent advantage of the epithermal supply at a pulsed spallation source for its extended Q -range.
4. Diffuse scattering surveys are well suited to single-crystal instruments at pulsed sources (array or 2-D detector) to display planes in reciprocal space; however, small crystal diffuse scattering studies, especially near Bragg peaks, due to coherent precipitates and atomic clustering and size mismatch are best done at a high-flux reactor in the triple-axis mode set to the elastic channel (filtering out phonons).
5. Residual stress measurements can be done on either source although strain mapping is still more efficiently done at a reactor.
6. Time-resolved studies depend largely on the time scales currently involved and the Q ranges covered and can be done at either source.
7. For diffuse modes in quasielastic scattering (rapid ion conductors, liquids, orientational diffusion) a reactor is superior for specific Q 's. For covering a wide range of Q (e.g., in viscous fluids), a spallation source may well be preferred.

3.2.4 New Opportunities with a Tenfold Higher Flux Source

We envision a tenfold higher flux source to offer opportunities for the following:

1. Residual stress volumes of 1 mm^3 could be routinely probed.
2. It would become possible to build a practical instrument for high-Q-resolution ($Q_{\min} < 1 \times 10^{-4} \text{ \AA}^{-1}$) SANS. For example, this would permit creep cavitation observations in bulk ceramics and metals along with other studies alluded to earlier.
3. High-resolution diffuse scattering near Bragg peaks ("Bragg SANS") would be feasible.
4. Earlier detection of damage in metals and ceramics would be possible.
5. The nucleation and growth stages of precipitation, especially in dilute alloys where the signal is weak, could be seen.
6. Neutron Brillouin scattering on glasses and other new materials (i.e., low-Q vibrational excitations might be probed).
7. The possibility of high-Q data collection with good Q-resolution at small Q (i.e., for liquids and glasses, would open up).
8. Small crystal and powder sample studies would include:
 - a. structures.
 - b. excitations.
 - c. diffuse scattering.
 - d. environmental studies.
9. Time-dependent possibilities:
 - a. in-situ studies matched to the kinetics (seconds or minutes).

- b. quasielastic analysis of diffuse scattering from both glasses and crystals, including orientational order, rapid ion conductors, H-metal systems, etc., requires a new regime of 1-10 μeV resolution to identify the molecular or atomic nature of various motional mechanisms associated with glass transitions.

3.2.5 Short-Term Needs

To make better use of existing facilities, and looking forward to neutron research needs when the next-generation neutron source comes on line, we list below shorter term needs for new and improved instrumentation. A very significant improvement to existing conditions would be to fund longer running cycles and upgrades on existing facilities. There are currently many high-quality proposals for research rejected because of time constraints. It would benefit both existing and future neutron facilities if there were:

1. An immediate upgrade of the power of HFBR at BNL from 30 MW to 60 MW. This would permit not only more experiments but experiments that currently cannot be done. This is a particularly high priority because of its cost-effectiveness and scientific impact.
2. Support for a strong program to develop 2-D detectors with a large dynamic range. For SANS, for example, the wide dynamic range of the experiment requires a 2-D detector with a very linear response to take advantage even of present fluxes.
3. For powder diffraction and glass research, the availability of additional detectors on spallation sources and multidetector powder diffractometers at reactors would represent a significant advantage.
4. New cold source technology to increase the flux of long-wavelength neutrons, coupled with improved small-Q (Bonse-Hart-type) cameras for SANS would be very desirable.
5. Development of a dedicated residual stress and texture spectrometer.
6. As ancillary equipment, the development of high pressure (15 GPa) at high temperature (1000°C) would be useful, and the development of furnaces operating above 2000°C.

7. A facility for the magnetic and gas-jet levitations of liquid samples to allow containerless studies of supercooled fluids such as liquid metals and liquid silica.
8. An accelerated development program for supermirrors as thermal guides.

3.2.6 Reference

1. G.H. Lander and V.J. Emery, *Nucl. Instrum. Methods B* **12** (1985), 525-561.

3.3 Chemistry

A.K. Cheetham (University of California - Santa Barbara)

J. Eckert (LANL)

D.A. Neumann (NIST)

H.L. Strauss (University of California - Berkeley), Chairman

3.3.1 Introduction

Neutrons provide a number of unique tools for chemistry. The first of these involves diffraction methods used to determine the structure both of individual molecules and of complex molecular assemblies. In many cases, the uniqueness of the neutron methods lies in the particular distribution of neutron cross sections across the periodic table and among different isotopes. This distribution, that differs markedly from that of x-ray cross sections, allows one to pick out the diffraction effects of particular atoms. The second set of neutron tools are inelastic scattering methods which probe the dynamics of the molecules and arrays. Often these methods are especially sensitive to the same atoms that were highlighted in the diffraction experiments.

A number of themes recur in the following examples. The availability of higher neutron fluxes would allow experiments on smaller samples or over larger energy or momentum ranges. Small samples are often the only ones available due to the difficulties and expense of synthesis, the problems of having a large sample under extreme conditions, or both. Alternatively, a sample changing with time can be followed more rapidly, or fixed samples examined at higher resolution. Often, this suggests the use of reactor sources at low energy and momentum transfer and the use of spallation sources at high energy and momentum transfer, but the optimum source depends on the details of the experiment. The reactor and spallation sources are complementary.

We present a number of areas of chemistry to which neutron methods have made seminal contributions within the past few years and that we feel will continue to be of importance in the future. Opportunities made available by advances in instrumentation are discussed by area.

3.3.2 Liquids and Solutions

A good example of some of these points is the study of liquids and solutions such as NiCl_2 in water where the cation forms complex ions, for example $\text{Ni}(\text{H}_2\text{O})_6$.¹ Since the neutron cross sections for H and D are comparable to those for the other ions, diffraction can determine the Ni-H (Ni-D) radial distribution function as well as all the other possible distribution functions such as O-O. This is in contrast to x-ray diffraction, which is relatively insensitive to H (or D). For liquids such as molten salts — molten NaCl, for example — neutron scattering provides the possibility of determining the various distribution functions individually — the Cl-Cl function, for

example — by examining systems containing different concentrations of the two Cl isotopes. Furthermore, the isotropic nature of the neutron form factor yields the radial distribution functions directly. This makes neutrons the method of choice for comparison with molecular dynamics simulations. It should be emphasized that such simulations make it possible to understand ever more complex systems. Data to calibrate the intermolecular force fields used in the calculations and to keep the calculations honest are vitally needed.

3.3.3 Energy- and Information-Related Materials

Hydrogen remains the fuel of choice for the future as oil reserves are being depleted, and metal hydrides represent the preferred way of storing hydrogen as fuel both on safety and energetic grounds. Neutron scattering techniques have provided much of the microscopic detail of hydrogen location and motions (both diffusive and vibrational) in metals and alloys including those of technological importance such as FeTi and LaNi₅. Some of the most fundamental work, which has come on very dilute samples for which high source intensity is a necessity, has, for example, determined the details of trapping of hydrogen in Nb at impurity sites of either substitutional metals or N or O impurities. Another outstanding example of the power of neutron scattering techniques is recent work using quasielastic coherent neutron scattering from D, which probes the correlations between the diffusive motions of the D atoms and the displacement fields these produce. The perceived importance of research in this area seems to depend somewhat on the energy supply situation, and may therefore be expected to increase dramatically as hydrogen becomes a necessary fuel.

Solid crystalline systems that allow ion conductivity are useful as electrodes for high-performance batteries. Examples are sodium beta-alumina, in which the sodium ions conduct; silver iodide, in which the silver ions conduct; and cubic stabilized zirconia, in which the oxygen ions are mobile. Neutron scattering has been used to define the structure of such crystals, and quasielastic neutron scattering has been used to show that a particular ion moves. There are also a number of materials that form 2-D (layered) structures with an intercalated ion able to move in the plane between layers. TiS₂ intercalated with mobile Li is such a system. In order to determine some ions that have a small cross section, it is extremely useful to have a source with increased flux. This additional flux would also allow the determination of correlations between the motions of the diffusing particles through the use of polarized beams.

Crystals with nonlinear and often anisotropic optical properties are useful for mixing and doubling optical beams and for forming optical waveguides and other devices. A particularly versatile series of compounds is based on KTiOPO₄ (KTP). In this series of compounds, the K may be replaced by Ag, Rb, Na, or H; the Ti, by Sn or Ge; and the P, by As. The parent material itself can be used as a frequency doubler from the infrared (Nd-YAG) to the visible. The index of refraction can be varied by changing the ions, and so for example, RbTiOPO₄ (RbTP) has a lower index of refraction than KTP. A waveguide for visible light can be made by cladding a rod of RbTP with KTP. These interesting and varying optical properties make it important to characterize both the pure materials and their mixed crystals using, in most cases, powder neutron diffraction.

$\text{K}(\text{Ti},\text{Sn})\text{OPO}_4$ systems are particularly easy to characterize since Ti has a negative neutron scattering length and Sn a positive one, leading to a large contrast between the various crystals.² Many challenges can be tackled in the future. An increasingly important optical material is $\beta\text{-BaB}_2\text{O}_4$ (BBB). Here, neutrons could determine the positions of the O even though the Ba is of much higher atomic number. However, the ^{10}B will absorb neutrons making it impossible to use a large sample. The experiment could be done, however, with a diffractometer of an order-of-magnitude higher flux than currently available, since this would allow the use of a much smaller crystal.

3.3.4 Catalysis

One of the major highlights of the 1980s has been the contribution of neutron scattering to our understanding of heterogeneous catalytic processes in microporous solids such as the aluminosilicate zeolites (Fig. 3.1). These important catalysts, in which the selectivity is controlled by the shape and dimensions of the pores and windows, are used for a wide range of hydrocarbon conversions in the petrochemical industry, including gasoline manufacture and xylene isomerisation. Both elastic and inelastic scattering methods have been applied, and the active participation in such work of scientists from many major companies, including Amoco, Chevron, DuPont, Exxon, Mobil and Union Carbide, attests to the industrial importance of the field.

A detailed knowledge of the crystalline structures of these materials is essential to a proper understanding of their catalytic behavior, and the unique properties of the neutron have facilitated precise refinements of the architectures of a number of important systems (e.g., Table 3.1). Both the sensitivity of neutrons to the light atoms (Si, Al, and O) that constitute such zeolite frameworks, and their ability to differentiate between silicon and aluminum, make neutrons superior to x-rays for such studies. The sensitivity to light atoms is strikingly illustrated by the direct observation³ of Brønsted acid sites in lanthanum zeolite-Y, which is an important component of the cracking catalysts that are used in gasoline manufacture. The difficulty of making single crystals of sufficient size and quality to perform x-ray measurements has been an important factor in such work, since it is widely recognized that profile analysis of powder data by the Rietveld method yields better precision when carried out with neutrons rather than x-rays.

The success of studies on zeolite structures has naturally lead to more demanding experiments that probe the behavior of adsorbed hydrocarbon molecules inside zeolite cages. Both the structure and dynamics of the sorbates are known to play a central role in zeolite catalysis. In this context, neutron diffraction measurements have established the precise location of benzene and pyridine molecules in the cavities of both Na-zeolite Y⁴ and K-zeolite L,⁵ and complementary studies by inelastic and quasielastic scattering, and nuclear magnetic resonance, have been used to elucidate their vibrational and diffusional properties. The Q-dependence of the quasielastic scattering is particularly powerful in differentiating between local and long-range translational motion. Together with state-of-the-art molecular dynamics simulations, such studies are shedding new light on the factors that control reactivity and selectivity in zeolite catalysis.

FIGURE 3.1 The location of pyridine in the catalyst, potassium zeolite-L, determined by neutron diffraction.³ The nitrogen of the pyridine forms a Lewis complex with potassium, while the aromatic ring enjoys short-range interactions with the aluminosilicate framework.

TABLE 3.1 Examples of Zeolite Structures Studied by Powder Neutron Diffraction^a

Zeolite	Reference
Ca-A	Adams <i>et al.</i> , <i>J Solid State Chem.</i> 68 (1987), 351.
TI-A	Cheetham <i>et al.</i> <i>Nature</i> 299 (2 September 1982), 24.
ZK-4	Eddy <i>et al.</i> , <i>Zeolites</i> 4 (1986), 449.
Na-Y	Newsam <i>et al.</i> , <i>J. Phys. Chem.</i> 90 (1986), 6858.
K-L	Wright <i>et al.</i> , <i>Nature</i> 318 (26 December 1985), 611.
KFI	Fischer <i>et al.</i> , <i>Zeolites</i> 6 (1986), 378.
rho	Fischer <i>et al.</i> , <i>J. Phys. Chem.</i> 91 (1987), 2227.
K-ferrierite	Pickering <i>et al.</i> , <i>J. Catalysis</i> 119 (1989), 261.
mazzite	Newsam <i>et al.</i> , <i>Mater. Res. Bull.</i> 20 (1985), 125.
ZSM-5	Taylor, <i>Zeolites</i> 7 (1987), 311.

^a Note that several of these systems are used primarily for ion exchange and molecular sieving applications, rather than catalysis.

Future challenges in this area will include the extension of the methods described above to new generations of shape-selective catalysts. These are certain to include 2-D systems, such as ue pillared clays (PILCS), and the recently-discovered *mesoporous* sieves⁶ in which the cavity dimensions are as high as 300 Å.

The impact of neutron methods in homogeneous catalysis has also been significant. In Ziegler-Natta reactions, for example, which are used in the manufacture of polyethylene and polypropylene, the nature and importance of agostic hydrogens, adjacent to titanium, has been revealed by single-crystal neutron diffraction methods.⁷ Inelastic scattering has also been used to probe the behavior of hydrogen in such systems, and it is now being used to examine chemical bonding effects in biological sensors based upon dihydrogen complexes.⁷ These metal-dihydrogen complexes may be viewed as arrested reaction intermediates in the oxidative addition of hydrogen to a metal. The details of the chemical bond formed between the dihydrogen ligand and the metal, and the attendant activation of the H-H bond, have uniquely been examined by neutron scattering rotational tunneling spectroscopy.⁹ This method is extremely sensitive to details of the barrier to rotation. The barrier of the dihydrogen ligand arises from the electronic interactions between dihydrogen and metal and is sensitive to the nature of the other ligands bound to the metal.

The availability of a more intense neutron source would facilitate the development of advanced tools to meet the challenges posed by the complexity of modern catalytic materials. In particular, both powder and single-crystal work would become feasible on much smaller samples (the availability of the large samples that are required for existing neutron facilities is a recurrent problem), and the interrogation of hydrogenous, rather than deuterated, powder materials would become a viable option. Furthermore, rapid *in situ* studies, such as are needed to monitor the

high-temperature transformation of zeolite precursors into high-performance ceramics⁶ (e.g., cordierite, which is used for catalytic converter monoliths) would come within reach. And the scope of Rietveld profile analysis would be further enhanced by the construction of an ultra-high-resolution powder diffractometer.

3.3.5 Hydrogen-Bonded Systems

The structural and dynamical details of hydrogen bonds (H-bonds) are of great importance in many areas of science such as biology, chemistry, and planetary geophysics. For example, the helical structure of biological molecules (DNA) is held together by H-bonds, the structure of liquid and solid water is dominated by H-bonding interactions, and ordering of the H in H-bonds is responsible for ferroelectricity. Much of the knowledge of H-bonded systems has been obtained by neutron scattering techniques, and this is likely to continue in the future. Single-crystal neutron diffraction remains the method of choice for an accurate determination of H-bond geometry, and inelastic neutron scattering (INS) can be made selectively sensitive to motions of the H-bond proton by isotopic substitution. Much of the current and future work on these systems centers on combining structural and vibrational data to derive an accurate form of the H-bond potential.

An excellent example of the many very basic questions on such systems that remain unsolved is provided by the recent work of Nelmes and collaborators¹¹ on KH_2PO_4 (KDP) and related systems. This study demonstrated, with the aid of high-pressure neutron diffraction techniques, that the isotope effect on the ferroelectric transition temperature is largely the result of differences in bond geometry and NOT primarily to differences in the quantum mechanical tunneling of H or D between the two minima of the potential. This conclusion has important consequences for understanding the dynamics of H-bonds as well as of proton transfer. Several other basic H-bond problems remain to be solved such as the nature of H-bonds in the limits of very short and very long bonds.

The actual tunneling motion of the H-bond proton in the double-minimum potential can be investigated with quasielastic neutron scattering, which provides spatial (Q-dependence) as well as temporal information on this process. The concerted exchange of H-bond protons in carboxylic acid dimers has, for example, been shown to be translational tunneling instead of a rotational motion of the two molecules, whereas the latter process holds for dimers of pivalic acid.

The question of the mechanism for the essentially undamped energy transport over long distances in biological systems is of enormous importance for the function of biological molecules and remains largely unsolved. This problem is being modeled with molecular solids that contain chains of H-bonds akin to the peptide chain in real biological systems. Prominent examples of such model systems currently under study by various groups are acetanilide and N-methylacetamide. These solids show a large number of anomalous vibrational bands in the optical spectra which have been attributed to the existence of nonlinear excitations. The latter were proposed to be vehicles for energy transport in biological molecules. H/D substitution is being used with INS to identify vibrational coupling with these molecular vibrations which may play a

role in this energy transduction process. Much higher source intensities will be absolutely necessary if such work, both structural and dynamical, is to be extended to macromolecules.

3.3.6 Fullerenes

Neutron scattering investigations have significantly added to our understanding of the structural and dynamical properties of many new compounds, the most recent example of which are the fullerenes and the compounds based on them. The prototype of this class of materials, C_{60} , displays great chemical versatility, reacting with alkali metals, halogens, free radicals, amino acid adducts, metal-organic complexes, etc. The potential for future applications of these fullerene derivatives seems promising when one considers the useful properties, including superconductivity and nonlinear optical effects, which have already been discovered.

For example, neutron scattering has supplied the most detailed information on the low-temperature structure of solid C_{60} , elucidating details of residual orientational disorder which escaped earlier x-ray scattering investigations. Resolution of these details is almost certainly made possible by the independence of the neutron scattering length on the momentum transfer. In contrast, the x-ray form factor decreases with scattering vector making it difficult to separate the decrease in Bragg intensity due to the residual disorder from that due to the thermal parameters. This residual disorder has not been predicted by any calculation of the orientational potential, indicating that none of the carbon-carbon interactions used in these calculations is adequate.

Above an orientational order-disorder transition at 260 K, coherent quasielastic neutron scattering on powder samples has been used to demonstrate that the rotational motion of the molecules can be well approximated by random-walk rotational diffusion. Below this transition, INS shows that the molecules librate with an energy of about 20 cm^{-1} . In addition to this information, neutron scattering measurements of the Q-dependence of the intensity have been used to determine the spatial character of these excitations. These measurements show that the orientational potential is stronger than that obtained from any calculation, yielding another important clue towards the development of an adequate carbon-carbon potential.

INS has also played an unusually important role in elucidating details of the intermolecular modes in C_{60} because of the unusually high I_h symmetry. This results in only 4 of the 46 distinct frequencies being infrared-active and 10, being Raman-active. Perhaps most importantly, neutron scattering measurements of the intramolecular modes in the superconducting compound K_3C_{60} have shown a remarkable broadening of some of the intramolecular modes with respect to those in pure C_{60} . This is direct evidence for strong electron coupling to these particular excitations, suggesting that they may be responsible for superconductivity in these compounds.¹²⁻¹⁴

The first measurements of the phonon dispersion curves have recently been made using a 3-mm^3 single crystal; however much more detailed dynamical information could be obtained if an order of magnitude increase in flux were available. For instance, x-ray diffraction measurements

indicate that there is a deviation from perfect spherical symmetry in the high-temperature phase. With more flux (or larger crystals), more detailed information on the spatial extent of the rotational motion could be obtained. The increased flux could also be used to increase the Q-range of experiments while maintaining an excellent energy resolution. This would enable the determination of the spatial character of the molecular motion observed by nuclear magnetic resonance (NMR) in the low-temperature phase of C₆₀. The improved flux would also make measurements possible on much smaller samples of the molecular derivatives being produced in a number of university and industrial laboratories. Here, neutron scattering could play an important role in determining the structure and the nature of the intra- and intermolecular forces which underlie possible applications. This information may, in turn, be used to tailor new materials for a particular purpose.

3.3.7 References

1. G.W. Neilson and J.E. Enderby, *Adv. Inorg. Chem.* **34** (1989), 195-218.
2. S.J. Crennell, J.J. Owen, A.K. Cheetham, J.A. Kaduk, and R.H. Jarman, *Eur. J. Solid State Chem.* **28**(2) (1991), 397-407.
3. A.K. Cheetham, M.M. Eddy, and J.M. Thomas, *J. Chem. Soc., Chem. Commun.* **1984** (1984), 1337-1338.
4. A.N. Fitch, H. Jovic, and A. Renouprez, *J. Phys. Chem.* **90** (1986), 1311-1318.
5. P.A. Wright, J.M. Thomas, A.K. Cheetham, and A.K. Nowak, *Nature* **318**(19) (26 December 1985), 611-614.
6. Mobil patent (1992).
7. M. Brookhart and M.L.H. Green, *J. Organometallic Chem.* **250** (1983), 395-408.
8. Z-W. Li and H. Taube, *Science* **256** (10 April 1992), 210-213.
9. J. Eckert, G.J. Kubas, J.H. Hall, J.P. Hay, and C.M. Boyle, *J. Am. Chem. Soc.* **112** (1990), 2324-2332.
10. R.L. Bedard *et al.*, Proc. Int. Zeolite Conf., Montreal, Canada (1992), in press.
11. R.J. Nelmes, M.I. McMahon, R.O. Piltz, and N.G. Wright, *Ferroelectrics* **124**(1-4) (1991), 355-360.

12. W.I.F. David, R.M. Ibberson, T.J.S. Dennis, J.P. Hare, and K. Prassides, *Europhys. Lett.* **18**(8) (1992), 735-736.
13. J.R.D. Copley, D.A. Neumann, R.L. Cappelletti, and W.A. Kamitakahara, *J. Phys. Chem. Solids* **53** (1992), 1353-1371.
14. K. Prassides, C. Christides, H.J. Rosseinsky, J. Tomkinson, D.W. Murphy, and R.C. Haddon, *Europhys. Lett.* **19**(7) (1992), 629-635.

3.4 Polymers and Complex Fluids

F. Bates (University of Minnesota), Chairman

C. Han (NIST)

J. Huang (Exxon Research and Engineering Co.)

E. Kaler (University of Delaware)

T. Witten (University of Chicago)

3.4.1 Recent History

Complex fluids, containing polymers, colloidal particles and/or surfactant assemblies, are distinctive because they have structures much larger than single atoms or small molecules. Despite their great practical importance in materials like plastic, rubber, and food and in processes like emulsification, lubrication, gelation, and coating, the understanding of complex fluids has until recently been chiefly empirical and Edisonian. However, the last decade has seen a dramatic expansion of scientific activity in complex fluids. This activity has shown the molecular basis for formerly mysterious macroscopic behavior. A large part of this achievement was made possible by advanced structural probes on length scales from tens to thousands of Ångströms. First, we outline the pivotal role that neutron facilities have played in this scientific advance. The following sections anticipate further advances that could be made with contemplated new facilities.

A development of major significance in complex fluids has been the observation of the distinctive molecular motions underlying the macroscopic behavior of the fluid. A central example is the molecular understanding of flow in a liquid of entangled polymers. Recently, the random motions of individual sections of such polymers were measured over a wide range of time scales, using the neutron spin echo (NSE) technique. The result gave striking support to one of the competing theories of this polymer motion. This experiment puts the understanding of stress and flow in polymer liquids on a new level of firmness. Related experiments have demonstrated the random motion of surfactant-laden oil-water interfaces in microemulsions, showing the importance of bending rigidity absent in unladen interfaces.

The miscibility of two chemically different polymers is a central concern in the design of polymer materials. It is crucial to understand the degree of immiscibility in a given blend and to characterize the space and time development of phase separation when it occurs. In the last decade, it has proven possible using neutron scattering to observe both the precursors of phase separation in marginally miscible blends and the kinetics of phase separation from its earliest to its latest stages. These measurements have confirmed and refined theoretical expectations and have allayed some persistent misgivings about them. At present, the miscibility studies are expanding rapidly, from simple model polymers to the random copolymers, which are important industrially. Neutron scattering is the probe of choice in these studies because of the strong and controllable contrast it offers and the wide range of length scales it probes.

Polymers and microemulsion interfaces are very deformable; thus, they have large fluctuations and respond strongly to changes in their environment. It is essential to understand how these molecular deformations occur in practice. In the 1970s, a central problem in this domain was resolved using neutron scattering. This was the characterization of the shape of a single polymer coil in an entangled melt of polymers. In the 1980s, the strong deformation of these polymers under various important conditions was shown. One such condition is created by stretching the polymer melt or a crosslinked rubber or gel derived from it. Recent neutron scattering studies have shown that the individual chains can deform much differently than the sample as a whole. Some parts of this distinctive chain deformation have yet to be explained. Strong deformation also occurs when polymers are attached to a surface by one end. Such grafted polymers are important in many complex fluids. In the last three years, the distorted state of grafted polymers has been explored in great detail via neutron scattering: several important regimes were identified, theoretical ideas were confirmed, and their practical limitations explored. The same was done for the analogous chain distortions encountered in block copolymer microdomains.

The fluctuating surfactant interfaces in microemulsions have also been characterized using neutron scattering. Here the sizes and shapes of droplets, the thickness of the surfactant layers, and the neutral mean curvature of symmetric, bicontinuous microemulsions were definitively measured. In these experiments, the controllable contrast afforded by a neutron probe was essential. Contrast variation also played a key role in determining the internal structure of both synthetic and biological surfactant micelles. SANS measurements have provided clear views of the nature and importance of both attractive and repulsive intermicellar interactions. SANS has also illustrated the importance and range of critical phenomena in micellar and microemulsion systems. Analogous studies on colloidal solutions have identified the conditions needed for stable dispersions of colloidal particles.

Neutrons have also proven advantageous in observing the microheterogeneity responsible for much of the macroscopic behavior of polymer fluids. A primary example is the microphase separation of diblock copolymers. Here, a wealth of new ordered structures controlled by shear flow have recently been identified, thus greatly expanding the potential for shaping materials on length scales of tens of nanometers.

In addition to these major scientific advances, neutrons have gained great acceptance as a routine probe for characterizing ill-defined materials such as gelified coal, refinery sludge, or complex food products. Neutrons scatter weakly, and their scattering contrast is controllable and predictable. Thus, they may be used to give unambiguous and overdetermined information about the sizes of structures in the unknown material. A noteworthy example from the last decade is the demonstration of fractal structure in a wide variety of colloidal aggregates important in separations, in coatings, in food products, and in ceramics. Another is the characterization of surfactant-stabilized colloids found, for example, in motor oil. Here, the dimensions and variability of the colloidal particles and their surfactant layers were definitively measured. The number of specific fluids being routinely characterized in this way is increasing rapidly.

These scientific advances have found a variety of significant technological applications. Discoveries about the microstructure and flow behavior of block copolymers have led to improved thermoplastic elastomers and pressure-sensitive adhesives. These technologies were pioneered in the U.S. The ability to predict the miscibility of copolymer blends has significantly increased the design capabilities in the rubber tire and polyolefin industries. An improved understanding of polymer conformations in solution has improved the development of oil additives. Knowledge gained in surfactancy is being used in detergent and emulsification products. Each of these represents large sectors of the chemical industry, which contributes a critical component to U.S. commercial competitiveness.

3.4.2 Future Applications

Recent history has clearly demonstrated the importance of neutron scattering in the study and application of polymers and complex fluids. Nonetheless the ultimate versatility and power of these techniques will not be realized without a significant increase in the available neutron flux. An order-of-magnitude or greater increase in flux in the U.S. would bring both evolutionary and revolutionary advances. Evolutionary developments are anticipated in nearly every type of experiment currently conducted where flux is the limiting factor. Qualitative advances, especially in examining time-dependent phenomena, would be possible. Such time-dependent phenomena are especially important in polymers and complex fluids, where many industrial processes (e.g., extrusion, drying, flocculation, or adsorption) occur on time scales between one and 100 seconds — just beyond the capabilities of present-day facilities.

One may also anticipate more dramatic, revolutionary, developments to accompany the increased flux, as they have in the past. For example, one expects to be able to study low-concentration systems like biosynthetic proteins, adsorbed polymers, and polyelectrolyte solutions in ways not previously accessible. Likewise, the increased flux will permit contrast-matched studies now impossible because of low signal-to-noise.

In the following sections, several representative scientific and technological examples are presented. These have been selected from an extensive list of exciting opportunities and should not be viewed as comprehensive. Given the growing importance of neutron scattering to our industrial competitiveness, we fully expect an advanced neutron source to support and spawn a broad complement of technologies in this area.

3.4.2.1 Kinetics

With an increase of ten times more in flux over current U.S. neutron sources, many important issues that involve the time-dependent measurement of structure factor can be addressed. Very often these subjects have direct implications for manufacturing, because a one-second SANS measurement could access the transient structural rearrangements that occur under actual

commercial conditions. These time-dependent processes can be divided into physically and chemically induced phase transitions and transformations.

Physically induced phase transitions are often driven by rapid temperature or pressure quenches, with subsequent changes that are governed by spinodal decomposition or nucleation and growth kinetics. Establishing the global relationships between structure, time, and properties requires a detailed determination of the time-dependent structure factor for a wide range of materials. Conducting such experiments in real time would be made feasible by an advanced neutron source. Such investigations are particularly important for commercial application when combined with flow and deformation, which distort the polymers and influence the thermodynamic phase boundaries. A major fraction of the polymer business, the largest sector of the U.S. chemical industry, is directly influenced by these issues.

The study and understanding of microstructure in many classes of materials such as block copolymers, surfactants, and colloids under static conditions has been a major part of the growth in SANS in the past decade. A challenge for the future is to understand the kinetics of formation and/or dissolution of these microstructured materials. For example, microphase-separated block copolymers are highly susceptible to orientation through deformation. SANS studies have shown that shearing or extensional flows can transform an ordered, but macroscopically isotropic, specimen into a highly aligned state with long-range order. However, the underlying mechanisms responsible for microdomain rearrangement are obscure. Rheological measurements demonstrate that, under commercially relevant processing conditions, domain relaxation occurs on the order of several tens of seconds. Applications of oriented block copolymers as anisotropic nanocomposites have not been realized to date due in part to a poor understanding of these time-dependent processes.

Many surfactants also order in water-forming bicontinuous, lamellar, cylindrical, and spherical microstructures. Shear alignment plays an important role in the application of these interesting soft materials due to the associated anisotropic viscoelastic properties. For example, some surfactants form long, rodlike or threadlike micelles in water. Such micelles entangle, and the rheology of aqueous solutions of these micelles is complex. Solutions containing less than a few weight percent surfactant are viscoelastic and display apparent viscosities as high as 1000 centipoise. Such rheological properties are useful in personal care and cosmetic products.

Because of the entanglement of the micelles, the solution is isotropic even though the individual micelles have an extremely high aspect ratio. In order to study the internal structure of and interactions between the micelles, they can be aligned in a shear flow field. Analysis of the scattering from aligned samples gives information about micelle cross section and persistence length, as well as intermicellar interaction potential. The relationship between the reptation time of an unbroken micelle and its average lifetime governs the rheology, and the connection can be probed by kinetic (stress relaxation) experiments.

As is the case for block copolymers, time-dependent phenomena that derive from flow or phase transitions, or both, will be made possible by an increased flux. Contrast matching and

low-concentration measurements will also be enhanced. In addition, beam geometry and wavelength distribution can be narrowed significantly to reveal fluctuation effects by diffraction linewidth analysis. While such experiments could in principle be better accomplished with synchrotron x-rays, the penetration of materials by neutrons makes feasible the design of sturdy deformation devices that are necessary when dealing with these highly viscous and elastic materials. Applications of surfactants include oil recovery, lubrication, emulsion stabilization, and health care products.

Often, phase transitions are induced by chemical reactions, which in combination with the aforementioned physical driving forces lead to a complex balance between chemistry, physics, and engineering. A representative example is reactive processing of toughened engineering plastics. Mixtures of two thermodynamically incompatible polymers are mixed and compatibilized by allowing the constituents to react at the evolving interface to form graft or block copolymer. During this process, the number and type of components change leading to drastic shifts in phase behavior and interfacial properties (see below). In order to design such materials, real-time experiments conducted during the processing step are required.

3.4.2.2 Dynamics

To understand the fundamental processes that govern the properties and behavior of a complex system, two basic questions need to be addressed: what is the equilibrium structure of the system of interest, and how does it respond to external perturbations? Once the structure is known, then it is possible to study how and why a system behaves the way it does. For polymer and complex fluids, the characteristic length scale is in the range of 1 to 1000 nm, and the corresponding characteristic molecular time scale is typically in the range of 1–1000 ns. It turns out that dynamics in this time scale are very difficult to access. NSE spectroscopy which utilizes the precession of the neutron spin in a magnetic field is a unique high-resolution technique particularly suitable for this investigation. Furthermore, by contrast variation schemes, the appropriately labeled components can be analyzed. For example, the diffusion of polymer chains in the highly entangled melt state is qualitatively different from that of smaller molecules. A theory that described the motion of a polymer chain in the melt was first proposed over twenty years ago. However, direct observation of this new diffusion process has proved to be very difficult. Important progress has been made recently by NSE experiments on a system consisting of dilute center-labeled chains (with deuterium) in the melt of other unlabeled, but otherwise identical, polymer. Many interesting scientific and technological issues of polymer systems are addressed in terms of the dynamic properties. Examples include adhesion phenomena and the rheological response of polymer melts.

In spite of the unique capabilities of the NSE technique, application of the current neutron spin echo device is mainly limited to the study of systems with strong scattering amplitudes. Even then, a typical experiment takes a week to ten days of beamtime to perform at the ILL, whereas a typical SANS experiment requires only hours to complete. The relative small number of experiments that can be scheduled in a given reactor cycle is one of the reasons that NSE has not

enjoyed the broader subscription that it deserves. An order of magnitude increase in the cold-neutron flux expected from an advanced neutron source will not only substantially increase the number of experiments performed, but more importantly, it will allow qualitatively different systems and a broader range of problems to be investigated. Examples include membrane elasticity on microscopic scales, diffusion of molecules in porous media, protein conformation fluctuations and dynamics of gels and cross-linked networks. Potential industrial applications will range from better design of controlled release of medicine and fertilizers, more efficient dispersant molecular architectures, stronger composite materials, and better adhesives.

In addition to NSE, which covers mainly the lower end of the relevant frequency spectrum for complex fluids, we also point out the importance of high-resolution TOF and backscattering instruments for the study of glassy states and molecular motion of the surfactant or lipid molecule on interfaces. These could have broad impact on a variety of industrial processes.

3.4.2.3 Polyelectrolytes and Biopolymers

Many of the polymers encountered in food, in pharmacology, and in other water-based fluids are electrically charged. These "polyelectrolyte" solutions have vastly different properties than do neutral polymers. Yet, the altered molecular conformation responsible for this difference has remained a mystery despite two decades of experiments and competing theories. This situation is due to the insufficient intensity of existing neutron scattering beams, as polyelectrolyte solutions of interest are intrinsically more dilute than those of neutral polymers. Given the results of attempts to "see" polyelectrolytes at existing facilities, it is clear that an order of magnitude increase in flux would be sufficient to give the clear-cut structural information needed.

Protein conformations and protein-protein interactions can both be probed by SANS, and information about both is essential to the design of improved protein separation and purification schemes. Separation and purification remain the costliest steps in producing proteins, especially at the level of purity demanded for therapeutic use. Annual sales of such products will exceed several billion dollars in 1992, so there is a substantial demand for quantitative design methods for protein separations.

The foundation of a rational thermodynamic approach to protein separations is knowledge of the potential of mean force acting between the proteins in solution. At the colloidal level, this potential can be measured and parameterized in ways similar to those used previously for micellar or microemulsion structures. Such measurements of the potential of mean force can also be compared directly to realistic computations and simulations. At the level of an individual protein, SANS can give information to low resolution (ca. 5 Å) that complements the information given by the crystal structure, when it is known, or provides a picture of the conformation when the crystal structure is unknown.

The instrumental needs are for high flux at all scattering vectors (Q) for measurement of dilute proteins and for contrast variation experiments. Resolution at low Q is critical for characterization of concentrated protein solutions, both to determine accurately the structure of any dimers or higher order aggregates that could form, as well as to determine the isothermal compressibility of the solution. Typical bioproduct streams contain less than 1% of the target protein, and current neutron fluxes do not allow quantitative measurements from such dilute solutions. Proteins are delicate and are susceptible to radiation damage, so high flux SANS measurements offer advantages over synchrotron experiments.

3.4.2.4 Interfaces

Most uses of complex fluids and polymeric materials are influenced by interfacial phenomena. Lubrication, adhesion, colloid stabilization, thrombosis in biomedical implants, and wetting are just several examples of physical processes that occur in commercially important applications. Interfacial science and engineering will play an even greater role in future industrial developments as our ability to manipulate materials at more microscopic length scales continues to evolve. Neutron scattering techniques will constitute an important tool in the design and exploitation of the resulting products.

Adsorption phenomena contribute to a wide range of industrial products and processes that will be influenced significantly by an advanced neutron source. Neutron reflection is a uniquely attractive technique for exploring how polymeric, colloidal, and micellar solutions interact with solid, liquid, or gaseous interfaces. Because thermal and cold neutrons penetrate many common materials (e.g., quartz and silicon), reflection measurements of interfaces can be conducted by passing the neutron beam directly through macroscopic pieces of solid or volumes of liquid. This is not possible with x-rays. Thus, absorption of polymers onto a solid substrate can be directly examined.

Many industrial processes rely on adsorption phenomena. Water-based (e.g., latex) paints are stabilized through the addition of surfactants to emulsion-polymerized polymers. Application and drying of such coatings relies on a delicate balance between particle repulsion in solution, and particle fusion upon drying. (A leading U.S. latex manufacturer currently makes use of neutron scattering to aid in the design of advanced emulsions.) As the nation moves towards a solvent-free coating industry, the ability to optimize latex stabilization will determine commercial success or failure. Proteins, platelets, and other blood constituents interact and adsorb on surfaces in ill-defined ways. Understanding these steps is crucial in developing biocompatible medical devices, a technological goal that has eluded us for over two decades. Polymers and surfactants are also used in detergents, as flocculants in water purification, and in separation operations during biosynthesis. In each of these industries, the relationships between adsorption and molecular conformation are poorly understood.

Application of neutron reflection to the determination of polymer adsorption onto quartz from solution has recently been demonstrated. However, these experiments were conducted using

relatively short polymer molecules in order to increase the polymer concentration near the liquid/solid interface. Such measurements on many practical systems would require prohibitively long counting times. Increasing the neutron flux would have two beneficial effects on these experiments. Firstly, the increased counting rate would make numerous adsorption studies feasible. Polymer chain conformations and surface coverages could be determined for polyelectrolyte and protein solutions which typically adsorb at low concentrations. Secondly, real-time surface dynamics will be observable. Macromolecules frequently attach and rearrange on surfaces relatively slowly, on time scales that may take several minutes or longer (e.g., thrombosis). The construction of an adsorbing layer could be followed in real time with a large increase in neutron beam fluxes over that currently available.

Polymer adsorption and rearrangement also plays a critical role in the processes of adhesion and wetting. Current research has focused on examining the static structure of polymer-polymer, polymer-solid, and polymer-gas interfaces using neutron reflection measurements. Long counting times exclude the possibility of directly probing how these interfaces respond to transient deformation. The molecular relaxation times of bulk polymers is relatively long. Thus, the dynamical window for cooperative surface rearrangements would be quite accessible with the increased fluxes contemplated here. An exciting prospect will be measurements performed on specimens held between moving parts. This will closely approximate the events that occur under industrial applications of adhesives and lubricants where transient structural rearrangements of microstructures occur. Current instrumentation simply cannot provide such temporal resolution.

Interfacial development in polymer blends and composites is often improved by the addition of copolymer, or through the creation of these copolymers via chemical reaction during blending. Valuable, and otherwise inaccessible, information regarding the evolution of interfacial structure with time can be obtained using neutron reflection measurements. Beginning with layered specimens with sharp interfaces, the combined effects of interdiffusion and chemical reaction could be unraveled. Such measurements can guide quantitative design criteria for optimal reactive blending — e.g., molecular weights, grafting or block architecture, and concentration. At present, reactive blending and interfacial stabilization using block and graft copolymers constitutes an annual multibillion-dollar U.S. enterprise that is practiced as a "black art."

Until recently, neutron reflection experiments have extracted only a small fraction of the information potentially available from this method. Due to flux limitations, attention has mainly focused on specular reflectivity. This reflectivity probes the average composition profile normal to the surface plane. In-plane structural features, which are manifested in the off-specular component, generally produce a significantly weaker signal that is often difficult to capture or interpret. Important applications of off-specular scattering can be found in micro- and macrostructured thin films. Langmuir-Blodgett and block copolymer films are two such examples. (Here we note that the pioneering studies of block copolymer thin films by neutron reflection were conducted by scientists from a leading U.S. computer manufacturer.) These materials are currently under investigation for use as coatings for disk storage devices, as photochemical resists, and as nonlinear optical materials. These self-assembling systems may be induced to undergo phase transitions or structural rearrangements (e.g., upon heating), thereby facilitating processing or enabling pattern formation. Issues related to phase transition dynamics and ultimate dimensional

and structural stability can be examined by a combination of specular and off-specular neutron reflection. With present neutron sources, the off-specular component cannot be fully exploited. A particularly attractive feature associated with using neutrons is the absence of beam damage to these delicate materials.

3.4.3 Source Considerations

Advances in experimentation of the type described in the previous paragraphs will be driven in large part by the availability of significantly greater neutron fluxes. A quantitative comparison of spallation versus reactor sources is not possible in the context of this brief report, although several technical trade-offs can be identified. A pulsed neutron beam affords a greater dynamic range of momentum transfer, Q , with relatively narrow Q resolution. Comparable Q ranges can be realized with a monochromatic reactor-based beam but require the use of multiple detectors or detector movement. Monodisperse beams ($\Delta Q/Q \sim 1\%$) are readily achievable but with a severe sacrifice in flux. In certain SANS experiments, high intensity at low Q is the most critical feature and, in these instances, reactor sources may hold some advantage. Overall, most of the projected research could be accomplished using either source, *provided the time-averaged beam intensity is sufficiently high.*

Two important factors in considering sources are predictability and reliability. Here, reactor-based sources seem more desirable. The design and construction of a reactor will be accomplished using demonstrated technologies and will operate with a high degree of reliability during scheduled operating periods. Many of the developments in SANS and neutron reflection will involve time-dependent phenomena (e.g., reactive blending, phase separation dynamics, etc.) that will depend on a steady, uninterrupted neutron beam. Currently, pulsed source neutron beams operate with frequent interruptions that make transient experiments difficult. Another concern lies with the instrumentation (e.g., detector) developments necessary to allow utilization of the extremely intense bursts of neutrons that will be generated. According to instrumentation experts, the anticipated neutron fluxes from an advanced steady-state source can be used with predictable improvements in detector technology.

We find compelling reasons to pursue both neutron source options. An advanced pulsed source would provide an avenue for new and innovative instrument development. An advanced reactor source represents a predictable replacement for our aging research reactors that would best serve the needs of the polymers and complex fluids community. Based on the urgent technological applications of these materials, we strongly urge that a reactor-based steady-state source be given first priority, with the companion recommendation that an advanced pulsed neutron source also be designed and constructed in the U.S.

3.4.4 Interim Instrumentation

An advanced neutron source will create a plethora of opportunities for basic research and development in polymers and complex fluids in the U.S. In order to exploit this facility effectively, the community must carefully plan to expand the user base during the prolonged construction period. This is particularly urgent in the industrial sector where limited beam time and applicability chance restricted corporate participation. Specific recommendations include:

- *Recommission the ORNL National SANS instrument as a user-based facility.* The single largest number of users at an advanced neutron source will work with SANS instruments. In order to develop this community, significantly more beam time must be made available.
- *Develop advanced detectors.* Utilization of the intense neutron fluxes anticipated will require faster and higher resolution detectors.
- *Develop focusing optics.* SANS, neutron reflection, and NSE instruments will all be greatly enhanced by a more brilliant source.
- *Acquire and operate an NSE spectrometer.* This method will benefit enormously from a 10-fold or greater increase in neutron flux. Experience with this technique must be developed prior to operation of the advanced neutron source.
- *Develop polarization analysis capability at existing SANS instruments.*

3.5 Biology

D. Schneider (BNL)
 P. Timmins (ILL, France)
 J. Trehella (LANL)
 S. White (University of California - Irvine)
 G. Zaccai (ILL, France), Chairman

3.5.1 Update of the Scientific Case for Neutron Sources

In the summary of the Shelter Island workshop on scientific opportunities with advanced neutron scattering facilities (1985),¹ biological work is not treated separately but appears mainly in the chapters on small-angle scattering and crystallography (single crystal and protein) and briefly in the chapters on high-resolution and chemical spectroscopy. There is a chapter on scientific horizons in structural biology in the report of the Expert Meeting on the European Spallation Source (1992),² as well as discussions in the contributions of the working groups on high-resolution and molecular spectroscopy, large-scale structures (small-angle scattering), and crystallography (high- and low-resolution diffraction studies). In both reports, the emphasis is on limitations arising from low flux and inadequate Q resolution and range and on how advanced sources should improve matters.

The advantages of neutrons and their complementarity with x-rays and electrons for radiation scattering studies of biological systems were described in the early 1970s: (a) isotope effects (especially the large difference between the coherent scattering lengths of H and ^2H (D) and between their incoherent cross sections), which allow sophisticated labeling studies; (b) scattering amplitudes which are independent of atomic mass (the scattering pattern is not dominated by the heavier elements); (c) the negative coherent scattering length of H (which gives a coherent scattering length close to zero for the H_2O molecule); (d) a choice of wavelength from below $1\text{\AA}$ to above $10\text{\AA}$ associated with soft energies and negligible absorption in most matter, leading, on the one hand, to high penetration and negligible radiation damage in biological samples, which are usually made up of molecules folded by weak chemical interactions (of the order of a few kilocalories per mole), and, on the other, to easy to design sample containers for special environments (e.g., of temperature or pressure); and (e) the energy dimension provided by inelastic or quasielastic neutron scattering in the picosecond to microsecond range of temporal resolution (the appropriate range for the study of thermal motions in macromolecules) and the large incoherent scattering cross section of H atoms which makes such experiments possible in practice. These are almost ideal properties for a radiation to study biological systems, but there are two major disadvantages: the much lower flux than that of even conventional x-ray generators; and, for structural work, the high background due to the incoherent scattering of H (which is the most abundant element in biological samples).

There are different levels of biological structure, from atomic resolution to the organization of large cellular complexes, which are all important in understanding biological function. Since the 1970s and certainly in the last decade, the field of molecular biophysics has been dominated by the rapid expansion of x-ray protein crystallography for atomic resolution studies. As a result of advances in instrumentation and molecular biology, the field was opened up to a large number of groups prepared to put in a major effort to overcome the difficulties of crystallizing biological macromolecules and to develop new crystallographic methods to tackle large and complex structures. The richness of information in an atomic resolution protein or nucleic acid structure and the power of computer graphics in conveying this chemical information to the nonspecialist in an esthetic and useful way have certainly made the effort worthwhile. It is this large community of x-ray crystallographers that is now providing the impetus in the development of synchrotron radiation methods and that is well-positioned to take advantage of the complementarity of neutrons and x-rays.

The neutron community is much smaller. Nevertheless, neutrons have contributed significantly to our understanding of the structure-function relation at *different levels* of organization. A few examples: the location of H atoms involved in the enzyme mechanism of trypsin; a description of the hydration shell of proteins and nucleic acids; the location of functionally important water molecules in a membrane protein; the distribution of protein and nucleic acid in the internal structure of chromatin and of viruses; the location of individual proteins in the *E. coli* ribosome, and of the components of *E. coli* RNA polymerase and their interaction with DNA; descriptions of protein-detergent interactions in membrane protein crystals, lipid, and membrane protein structures; and the description of the dynamical transition in myoglobin and its correlation with protein function by INS.

Structural work at the molecular level is expected to continue increasing because of the progress in fundamental molecular biology driven by health concerns and the developing biotechnology effort. For a few years now, industry in the U.S. has been investing in the determination of macromolecular structures. Despite the wealth of information in an atomic structure of a macromolecule obtained by x-ray crystallography, it should be considered as a starting point in the understanding of a biological problem rather than an end in itself. Its limitations are that it is usually a partial structure (H atoms are missing, part of the structure may be disordered and will not appear in the density maps, the macromolecule may only be a small part of a larger interacting system) and that it is a time and spatial average over all the molecules in the particular environment of the crystal. Because of their special properties, neutrons can play an increasingly important role in contributing to the missing information, provided experimental and working conditions are improved sufficiently to create a new scientific impetus in the field. In the past, neutron studies have been limited also by sample preparation, in the case of the specific labeling of large complexes, for example. The advent of genetic engineering and progress in protein chemistry, however, now provide powerful methods for the controlled construction of biological systems and their production in the quantities required for structural study.

3.5.2 Reactors vs Spallation Sources

Biological studies make use of thermal and cold neutrons in elastic and inelastic scattering experiments over different energy and Q ranges (10^{-6} – 10^{-1} eV, 10^{-4} – 6 Å $^{-1}$, respectively) with different resolutions. Theoretical considerations lead to the conclusion that reactor sources would be preferable for certain experiments and spallation sources for others. There is wide experience on reactor sources for all classes of experiments mentioned above, which permits a quantitative extrapolation of what could be expected from improved sources and instrumentation. At present, too few biological experiments have been done at spallation sources to evaluate their potential accurately. Extrapolation from simpler systems in physics and chemistry is difficult because of the inherent signal-to-noise problems associated with biological samples. It appears, however, that reactor sources will remain pre-eminent for cold neutron applications, particularly SANS, a technique with applications in a wide range of biological systems, which attracts the most users. SANS in structural biology is carried out more successfully on steady-state and cold-moderated sources than on pulsed sources because the intense long-wavelength spectrum accessible in the steady-state provides the high- Q resolution necessary for the investigation of large biological complexes of characteristic dimensions of several hundred Ångströms. For high-resolution crystallography, however, spallation sources may be competitive if a time-integrated flux of about 10% that of a reactor source becomes available, although somewhat more complicated analysis will be required. Pilot experiments planned at LANSCE and ISIS should provide data for a quantitative comparison in the next few years. In the case of inelastic and quasielastic scattering experiments by TOF methods, reactor instruments provide the highest sensitivity at present, even though, in principle, spallation sources should be competitive. Sensitivity is important for biological studies because most experiments are difference experiments with low signal-to-noise ratios.

Considering the available data, we conclude that the molecular biophysics community would be best served by a reactor source rather than a spallation source.

3.5.3 Tenfold Increase in Flux

Since the signal-to-noise ratio in most biological samples is essentially sample dependent, a factor of 10 increase in flux on the sample would lead to shorter measurement times (with associated shorter alignment times and the possibility of measuring weak diffraction corresponding to higher orders) and/or the possibility of using smaller samples. This would lead to a dramatic increase in the number of biological problems that could be investigated by neutrons.

Probably the most important improvements would come in the fields of protein crystallography and inelastic scattering where experiments are, at present, almost invariably flux limited. In protein crystallography, the required crystal volume could be reduced to less than 1 mm 3 (at least for low molecular weight proteins) approaching the usual maximum for protein crystals. Combined with instrumental improvements such as bigger detector areas and sample-related improvements such as the production of genetically engineered fully deuterated proteins, this would open the field to more studies based on scientific interest rather than the availability of

samples of suitable size. In the field of INS, samples of ~100 mg are currently necessary. This is impossible to attain for many molecules of biological interest, whereas 10 mg is much more reasonable.

Where large crystals are available and for membrane or fiber diffraction, higher flux would lead to the measurement of higher orders of diffraction.

Even in the field of SANS, the flux gain will lead to projects currently excluded by low contrast and very dilute samples. It would have particular advantages for specific labeling of small components in a large sample matrix (e.g., chromatin or muscle fibers).

We should note that by optimizing instrumentation and methods at present high-flux sources, gains in measuring time and/or sample size of more than an order of magnitude can be envisaged — for example, by using perdeuterated samples, larger detector areas, and the quasi-Laue technique in protein crystallography.

3.5.4 Short-Term Goals

The most remarkable recent advances in biomolecular structure determination by neutron scattering were achieved at facilities that offer a full complement of scattering instruments operated and supported by an active local staff of research biologists. Structural biology is a multidisciplinary science that owes its rapid advances to a combined use of many scattering and spectroscopic techniques, as well as the prompt integration of new developments in biochemistry and molecular biology. Short-term support should focus on the strengthening of multidisciplinary, multi-instrument facilities.

3.5.4.1 Expansion of the Neutron Structural Biology Community

In order to take full advantage of powerful neutron sources for biomolecular research, it is imperative to expand greatly the neutron structural biology community. Despite the fact that there are significant and important problems of biology that can be addressed only by means of neutron scattering techniques, the number of investigators using neutrons is small and declining in the U.S. because: (1) the number of facilities has been limited; (2) existing facilities have not been easy to use; and (3) the main facility for neutron structural biology in the U.S., BNL, was not operating for a two-year period. To make matters worse, the ILL has recently shut down for repairs that may take two or three years to complete. Given this situation, it is clearly in the interest of the worldwide structural biology community to make a major effort to expand the capability of the U.S. for neutron structural biology. The main focus of this expansion should be in the area of protein crystallography.

The expanded neutron structural biology community must consist of principal investigators from universities, government laboratories, and private industry. The training of graduate and postdoctoral students is of obvious and central importance to this expansion. But, students cannot be attracted to the field unless they are confident that they can build careers based upon neutron scattering methods. This requires an adequate number of instruments of varying types whose future operation is assured. Thus, two issues arise as critical for the successful expansion. First, we must assure an adequate number of instruments which are routinely available to users inside and outside government laboratories. Second, training programs involving national and university laboratories, should be instituted. Cooperation with laboratories in Europe is highly desirable.

3.5.4.2 Critical Success Factors for the Expansion

The single most important issue for the successful expansion of neutron structural biology is the further development of strong independent research programs at the national laboratories with strong life science departments. Critical masses of independent principal investigators at these laboratories will assert strong pressure on the development of state-of-the-art instrumentation that is convenient to use. The existence of such facilities will in turn attract users from the university and industrial research communities. However, it is unrealistic to expect such facilities to evolve if the principal investigators at the national laboratories are burdened with instrument maintenance and user-support responsibilities. That is, not only must independent research programs be expanded, but the technical support teams for the instruments must be expanded as well. Thus, in the ideal world, the principal investigators would focus on the accomplishment of scientific goals and would, in the process, specify performance criteria for instruments constructed and maintained by the technical support teams.

3.5.4.3 Instrument Development

An on-going instrument renewal and modernization program is a key element to the reinvigoration of the neutron user community in molecular biology. Only sustained efforts to overhaul outdated equipment will assure availability of efficient and easy-to-use first-rate instruments. Continued development efforts can be carried out with minor interruption of instrument availability and, more importantly, will preserve talent and capabilities to develop first rate instrumentation for use at existing facilities, as well as on any of the new sources under consideration. Specifically, the power of the HFBR should be increased to its maximum allowable level, and the instrument renewal program proposed by BNL should be supported. The experimental SANS capabilities are generally good. However, the BNL SANS spectrometer that is used primarily by molecular biologists because of its favorable high-flux beam at long wavelengths (cold source moderated) is in need of a significant upgrade to bring it to the standard of the state-of-the-art instruments such as those at the ILL or NIST.

The capacity for protein crystallography should be significantly enhanced by the construction of additional diffractometers. The only protein crystallography instrument in the U.S.

is at BNL, and the waiting list for its use is long. Recent experience also shows that the protein crystallography station at BNL is easily adapted for work on partially ordered systems, such as fibers and membranes, so that if an adequate number of protein crystallographic instruments existed, they could be used for a wide variety of biological problems.

The use of the quasi-Laue technique should be carefully explored and, if successful, exploited. A productive neutron quasi-Laue diffractometer will have to be fitted with large area detectors that can intercept as much of the scattered intensities as possible. On a pulsed source, these would need to be electronic detectors, whereas on a steady-state source, these could be image-plate detectors behind converter screens. In either case, it is critical to step up detector development efforts. Research on multilayer neutron optical devices must also be developed. Instrumental developments at the ILL and BNL, as well as at LANSCE and ISIS, will allow an assessment of the gains to be obtained from the quasi-Laue method which is expected to improve data collection rates by a significant factor at either reactor or spallation sources.

At present, there is no scientific community in the U.S. active in the study of dynamics in biological macromolecules by INS. Appropriate TOF instruments will come on line at NIST in the next few years. It is strongly recommended that a special effort be made to develop biological projects on these instruments.

3.5.5 Summary

3.5.5.1 Update of Scientific Case

- Genetic engineering has led to the production of large quantities of material needed for structural studies. Specific deuteration, perdeuteration, and the production of designer molecules has become possible.

3.5.5.2 Tenfold Increase in Flux

- Will enable smaller sample volumes to be used, in particular, realistic crystal volumes, leading to a great increase in potential projects.
- The resolution (number of diffraction orders) can be extended.
- New experiments using dilute labels and low contrast will become feasible.

3.5.5.3 Reactor vs Spallation Source

- A reactor with a cold source is the preferred facility, due to the importance of cold neutrons and versatility.
- Pilot experiments should be carried out on spallation sources, in particular, to compare the use of the quasi-Laue technique with that on reactor sources.

3.5.5.4 Short-Term Actions

- Expand the user community through exchanges and training schemes.
- Upgrade existing facilities.
 - At BNL: Increase the power of the HFBR to the maximum allowable level. Construct a second crystallography station, a quasi-Laue station, and upgrade the SANS facility.
 - At NIST: Make the facilities more widely available to the biological community and, in particular, encourage the use of TOF facilities to generate a community of users interested in the use of INS to investigate protein dynamics.

3.5.6 References

1. G.H. Lander and V.J. Emery, *Nucl. Instrum. Methods B* **12** (1985), 525-561.
2. A.D. Taylor, ed., *Instrumentation and Techniques for the European Spallation Source*. Rutherford Appleton Laboratory, RAL-92-040 (Chilton, U.K., 1992).

3.6 Instrumentation

G. Felcher (ANL)
 C. Majkrzak (NIST)
 F. Mezei (HMI, Germany)
 D. Richter (KFA, Germany)
 R. Scherm (Physikalisch-Technische Bundesanstalt, Braunschweig, Germany), Chairman
 A. Taylor (RAL, U.K.)

3.6.1 Introduction

The ILL is without doubt the world's leading institution in the field of science performed with neutrons — investigation of condensed matter by neutron scattering as well as fundamental physics. What made the ILL a success?

- **The reactor** had been designed exclusively for beam experiments, including a hot and (today) two cold sources.
- **Instruments** have been designed for many different applications covering a wide range of energy and resolution. In an ongoing development program, devices and instruments had been continuously upgraded and renewed.
- **Open society:** The ease of access introduced neutron scattering as a tool for many users into a wide variety of scientific areas. This and the institute's international clientele made the ILL also a focus of scientific communication. (A similar philosophy had been adopted at the ORPHEE and HMI reactors and the pulsed source ISIS.)

The U.S. clearly had a leading role in neutron scattering up to the 1970s. The HFBR at BNL was the first high-flux reactor dedicated to beam experiments. Its instrumentation was excellent although narrowly focused. HIFR at ORNL excelled at the determination of magnetic structures using polarized neutrons. Later, pioneering work on TOF techniques at the pulsed source IPNS at ANL was the basis from which ISIS and LANSCE drew experience for their second generation TOF instruments. Today, we witness the growth of new instrumentation at the NIST reactor, and there, for the first time in the U.S., a guide hall has been constructed and an efficient hydrogen cold source will become operational soon. A number of instruments similar to the portfolio now common in Europe are being constructed.

The DOE operates two high-flux reactors. Instrumental development at these reactors has been rather modest in the past fifteen years.

We believe that there is an urgent need for the DOE to sponsor coherent long-term R&D in the areas of sources, devices, and instruments. We strongly underline the need for state-of-the-art cold sources, neutron guides, and associated instruments, e.g., SANS and submicrovolt-resolution spectrometers.

Such an instrumental development program will revitalize existing sources. Furthermore, it is a prerequisite for the efficient utilization of a next generation source.

For optimum utilization of an advanced source of either type, it is essential to maximize the number of available instruments. At a reactor, this is accomplished by cold neutron guides, allowing 10-15 instruments to be accommodated in place of one or two classical instruments at the reactor face. With some development, this concept can be extended to thermal neutrons. At a pulsed source, multiplexing is achieved with multiple target stations served by the one accelerator. Guides are also used, but instruments usually require an end-of-guide position. In this way, an advanced source of either type can support a suite of 30-60 instruments.

3.6.1.1 Comparison between Pulsed and Steady State Sources

The relative merits of each source may be evaluated in the simplest approximation by comparing the average flux of a reactor with the peak flux of a spallation source. In this sense, the ILL 10^{15} n/cm²·s flux is equivalent to the peak flux at ISIS (160 kW). Although the steady-state flux $\bar{\phi}$ at the ILL is greater than the peak flux $\hat{\phi}$ at ISIS at cold energies, the situation is reversed at higher energies.

To be more specific, one would have to compare two well-designed instruments, such as a two-axis diffractometer with a TOF powder instrument or TOF spectrometers on either source. The figure-of-merit is the peak flux times the time-utilization factor. The latter is the fraction of time during which the spectrometer collects relevant data. It includes factors such as the TOF frame length and nonoptimum duty cycles, as well as the portion of the scan which eventually defines the Bragg peak or classical phonon scan. In addition, crystal reflectivities have to be weighted against chopper transmissions, focusing crystals compared with converging neutron guides, etc.

Experience shows that pulsed sources have performed significantly better than was anticipated 8 years ago. A reactor and a pulsed source with equivalent peak flux complement each other. In Europe, the ILL (10^{15} n/cm²·s) is nicely complemented by ISIS (160-kW proton beam). Their instrument suites cover complementary areas with significant overlap (Fig. 3.2).

We believe that a 1-MW spallation source will be required to cover basically the entire field currently served by a 10^{15} reactor. In addition, it will provide scientific opportunities for neutron scattering which are unrealizable at a steady-state source. Scaling this up, the same comparison applies to a 5-MW pulsed source and ANS.

FIGURE 3.2 Complementarity of a Reactor and a Pulsed Source.

3.6.2 List of Instruments Worldwide

The following is a list of principal instruments for elastic or inelastic scattering which are currently available at laboratories in the U.S. and in Europe and Japan. As a reference, a state-of-the-art instrument from the ILL or ISIS is indicated by an asterisk (*). Technical issues pertaining to individual instruments as well as proposed improvements specific to both reactor and pulsed source are also addressed. A further section deals with the development of components and devices.

3.6.2.1 Elastic Instruments

a. SANS

World

D11, D22 * at ILL
5 in Germany
3 in France
Risø
ISIS
KENS
JAERI

U.S.

2 at NBSR, 30 meters
1 at NBSR, 8 meters
1 at HIFR
1 at HFBR
1 at IPNS (second almost complete)
1 at LANSCE

SANS instruments need high average cold neutron flux. In order to reach low Q , a long collimation path is required which limits the usable wavelength band. A D11-type instrument with 10% resolution at 8 Å, if transferred to a 50-Hz spallation source, would just fully utilize the time-averaged flux of this source. Shorter instruments at pulsed source will gain with decreasing length: a D17-equivalent instrument at 8 m would gain a factor of six over the average flux. Lowering the source frequency at constant pulse intensity does not change the situation. On the other hand, a pulsed SANS instrument offers better resolution ($\Delta Q/Q$) and a larger Q -range. SANS instruments at pulsed sources would greatly benefit from focusing devices, which effectively shorten the machine.

In addition:

- Double-crystal Bonse-Hart allows Q as small as 10^{-5} Å^{-1} . Improvements by multilammellar crystals are being developed.
- Further development of beam polarization and polarization analysis.

- Focusing techniques R&D under way.
- Real-time SANS may allow the study of kinetic processes.

b. Reflectometry

World

CRISP * at ISIS
 DESIR at Saclay
 KFA
 Risø
 Dubna

U.S.

1+ [1] at NBSR
 SPEAR at LANSCE
 HFBR
 POSY I and II at IPNS

By measuring the specular neutron reflectivity as a function of glancing angle from a surface, the chemical and magnetic density profiles normal to that surface can be determined. Neutrons are particularly suitable to study organic or hydrogenous films and multilayered structures such as diblock copolymers. Neutron reflectivity measurements are performed at both pulsed and steady-state sources. Direct comparison of the reflectivity curve of a diblock copolymer sample measured at NIST and LANSCE demonstrated that comparable results can be obtained on these two instruments. There are advantages and disadvantages to both techniques, but these are of a nonessential nature. The equivalence of the two methods extends to measurements of off-specular scattering which contains information on in-plane structure at surfaces and interfaces.

Increased source intensity would allow a new class of experiments involving weaker scattering density contrasts, smaller samples, and higher spatial resolution.

At the present time, the U.S. has state-of-the art instruments on both types of sources.

In addition:

- Polarized beams already exist (POSY-I, NBSR, and CRISP).
- Grazing incidence diffraction has been tested at NIST and the ILL (EVA). X-rays are usually preferred except for surface magnetism.

Possible Developments:

- Multiplex the incident beams.
- PSD for kinetic studies (proposal ANL).

c. Diffuse Scattering**World**

D7 * at ILL

(inelastic scattering is suppressed
with polarization analysis)

KFA

SPAN, E2 at HMI

SXD at ISIS

KENS

U.S.

No dedicated instrument

(triple-axis spectrometers or diffractometers
can be used)

Usually long-wavelength neutrons beyond the Bragg cutoff are required. Coarse energy analysis is used to discriminate against inelastic scattering. Great benefits can be obtained from polarization analysis. These cold neutron instruments can be implemented on both steady-state and pulsed sources.

Possible developments include efficient polarization analysis and extension to shorter wavelengths.

d. Powder Diffraction**World*****High resolution:***HRPD (resolution, 4×10^{-4}) * at ISIS

D2B at ILL

Risø

2 at KFA

HMI (stress analysis)

Saclay

U.S.HRNPD at HFBR (like D2B, but better
monochromator)

BT-1 at NBSR

SEPD at IPNS

GPPD at IPNS

NPD at LANSCE (stress analysis)

High intensity:

D1B * at ILL (kinetic experiments)
 D20 at ILL
 Polaris at ISIS (90° pressure cell)

HIPD at LANSCE (90° pressure cell)

The number of atomic parameters obtainable by powder diffraction was increased by a factor of 10 a decade ago, thanks to the development of Rietveld analysis and the higher Q-resolution obtained even with primitive TOF instruments. Although comparable resolution has been achieved at diffractometers on steady-state sources, pulsed source TOF instruments have an edge when one needs:

- an extended Q-range, as in the case of complex structures.
- a limited region of diffraction angles, as in a case of high-pressure experiments.
- an angle-dependent spectrum.

A very important case is the study of the strain resulting from the application of a stress. Steady-state diffractometers are important for long-d-spacing diffraction peaks, as in the case of complex antiferromagnetic structures.

Projections:

- Analysis of multiphase samples and their behavior across phase transitions.
- Application of other unidirectional forces (for instance, magnetic fields) that may alter the intensities more than the positions of the Bragg reflections.
- Development of tomographic methods for stress-strain analysis.

e. Single-Crystal Diffraction (chemical and magnetic)**World**

D9 * at ILL
 D19 at ILL (multidetector)
 Many 4-circle diffractometers
 SXD at ISIS
 E2 flat cone at HMI

U . S .

Several at HFBR
 NBSR (under development)
 SCD at IPNS
 SCD at LANSCE

2 of 2

Steady-state-source instrumentation is optimized for structural studies which require the accurate determination of (known) Bragg intensities; pulsed source instrumentation excels for wide reciprocal lattice surveys (incommensurate structures, diffuse scattering etc.).

Steady-state devices will benefit from monochromator development. Pulsed devices require further experience of λ -dependent correction. Multidetector technology is important for both types of source.

f. Protein Crystallography

World	U.S.
--------------	-------------

DB21 at ILL	HFBR
-------------	------

Quasi-Laue techniques hold much promise for protein crystallographic studies on both types of source. The TOF instrument has the advantage that it discriminates against incoherent background scattering from the many hydrogen atoms in the protein.

Instrumental development projects are anticipated at the ILL, ISIS, and LANSCE in this field.

g. Liquids and Amorphous

World	U.S.
--------------	-------------

SANDALS * at ISIS D4 at ILL	GLAD at IPNS (1/10 of SANDALS)
--------------------------------	--------------------------------

The rich epithermal spectrum from pulse spallation sources gives them several advantages for the study of pair correlation functions in liquid and amorphous materials.

- Broad Q-range ($0.1\text{--}100\text{ \AA}^{-1}$), leading to high real-space resolution.
- Inelasticity corrections are minimized by scattering at low angles. This is especially important in studying hydrogenous (and biological) systems.
- High count rates for isotopic substitution experiments. Here, detector and monitor stabilities better than 0.1% are required.

3.6.2.2 Inelastic Instruments

a. Collective Excitations

World

U.S.

Thermal

IN8 * at ILL
ORPHEE (double monochromator)
Risø
PRISMA at ISIS

H4, H5, H7, H8 at HFBR
BP1 at HFIR
BT-2, BT-4, BT-9 at NBSR

Cold

IN14, IN12 at ILL
Risø
ORPHEE
Berlin

HFBR (double monochromator)
on cold source
2 at NBSR

Hot

IN1 at ILL (hot source)
MARI at ISIS (better than TAS for
magnons >100 meV; good gain for
2-D problems >20 meV; essentially
for magnons)

No hot source
PHAROS at LANSCE (under construction)

Polarized

IN14 at ILL (cold source) *
IN20 at ILL (composite Heusler)
IN12 at ILL

BP1 at HFIR (^{57}Fe)
HFBR (Heusler, intensity = 1/50 of graphite)
BT-2 at NIST (Heusler)

Traditionally, TAS with thermal neutrons has been, and continues to be, one of the most important and successful efforts in condensed matter research in the U.S. and Canada. Nevertheless, comparable instrumentation for high-resolution cold neutron and high-energy spectroscopy is completely lacking at steady-state sources in the U.S. In these areas European facilities are far ahead. A first-rate cold source is desperately needed at one or more DOE neutron scattering centers. Although not fully proven as a substitute for conventional TAS techniques, TOF methods are being developed at pulsed sources and are worth pursuing. This applies particularly at energy transfers above 100 meV.

In addition to the obvious gains in performance which can be obtained with increased source strength, significant improvements can be made with better crystal monochromators. This includes the use of doubly-bent or composite anisotropic mosaic crystals and the development of bent crystals with lattice gradients. A more flexible choice of instrumental resolution will result from the development of magnetic monochromators utilizing polarized neutrons and energy-dependent spin-flipping devices (Drabkin). This device allows the energy line shape to be adjusted electronically and decouples the energy resolution from the angular divergency. An essential prerequisite in realizing this goal is the development of high-efficiency polarizers (reflectivity or transmission $> 80\%$) with high-spin contrast. Improved polarizers are also needed for TAS studies of magnetic excitations.

b. High-Energy Spectroscopy

World

MARI * at ISIS
HET at ISIS
eVS at ISIS

U.S.

HRMECS, LRMECS at IPNS
PHAROS at LANSCE (under construction)
PHOENIX at IPNS

The rich epithermal spectrum, high peak flux, good intrinsic resolution and signal-to-noise ratio allows chopper spectrometers at pulsed sources to:

- access high energy transfer (up to 2000 meV) at low associated Q for magnetic studies;
- access a wide range of Q- ω space for vibrational studies of polycrystalline and amorphous materials.

Together with resonance-based instruments on a pulsed source, chopper spectrometers can achieve very high Q for momentum density studies of quantum fluids and other low-mass systems such as hydrogen-bonded materials.

c. Thermal-Neutron Spectroscopy

World

MARI * at ISIS
TFXA at ISIS
IN4 at ILL

U.S.

HRMECS, LRMECS at IPNS
FDS at LANSCE
TRASH I + II at NBSR

TOF methods favor pulsed sources. Often, small values of Q are needed for form factor determinations and other reasons. Incident energies some 5-10 times ω , often at the higher end of the thermal energy range, are then required.

c. Cold-Neutron Spectroscopy

World

IN5, IN6 at ILL
IRIS at ISIS
NEAT at HMI
MiBeMol at ORPHEE
KFA
LAM80-ET at KENS

U.S.

QENS at IPNS
NBSR (medium resolution TOF)
NBSR (high resolution TOF; under construction)

- TOF spectrometers are ideally suited for pulsed sources. They effectively utilize the peak flux, modified by a time-utilization factor reflecting nonideal repetition rates. For cold neutrons, this factor is typically 1/2.
- Inverse-TOF spectrometers have a very large dynamic range (allowing, for example, moments to be determined), but lower backgrounds are required for quasielastic scattering. Further development is encouraged.
- There is a strong need for cold TOF machines at both reactor and pulsed sources in the U.S.

Projections:

- Spatial and time focusing promise significant intensity gains for all types of chopper spectrometers.
- Bunching techniques by accelerated crystals or Drabkin devices promise intensity increases.

c. High-Resolution Spectroscopy**World****U.S.**

IN11 * at ILL (NSE)

IN10 at ILL (backscattering) *

IRIS at ISIS

IN15 at ILL (NSE)

SPAN at HMI

KFA (backscattering)

NBSR (backscattering, in construction)

NBSR (NSE, under construction)

The backscattering spectrometer with its microvolt resolution as well as the spin-echo instrument with its nanovolt resolution have been very successful at the ILL. At a pulsed source, multiplexing the backscattering scheme (as proposed for SNQ) would utilize nearly the full peak flux of a strongly coupled cold moderator. Gains of the order of 10-15 are expected.

A spin-echo instrument at a pulsed source needs a long flight path. Similar to SANS, it needs only coarse wavelength resolution. Gain factors of 4-5 are expected with the benefit of increased Q-resolution and larger dynamical range (Fourier time $\sim \lambda^3 \times H$). The TOF spin-echo method is still to be proven.

Projections:

- Zero-field NSE should be developed; great potential for SANS and large area detectors.
- Inverse TOF with microvolt resolution and large Q-range is desirable.
- 4 neV resolution with backscattering using, for example, GaAs [111] crystals.
- At a reactor (NIST), phase-space transformation to increase flux of backscattering instrument.

3.6.2.3 Devices

a. Moderators

Hot Sources

They are needed in a reactor, but not at a pulsed neutron source, because the $1/v$ tail of the unequilibrated neutron spectrum gives an abundant supply of hot neutrons. At the ILL, a hot source feeds four instruments (D4, D3, D9, IN1). None exists in the U.S. ANS should have a hot source unless paralleled by an adequate pulsed source.

Cold Sources

Their importance has increased steadily in the past two decades.

In reactors, the standard moderator is now liquid H_2 or D_2 set near the core to optimize the flux. While in Europe every major nuclear center has one, in the U.S., only HFBR has a liquid H_2 source operating, retrofitted in a NON-optimal geometry. At NIST, a liquid H_2 source will be installed next year.

At spallation targets, cold sources are thought more as part of the individual beams. Thus, the source can be customized better for the requirements of the individual systems. A fair amount of development is expected, in regard to:

- Cooling material. Solid methane has been found to give a cooler spectrum than liquid hydrogen at the same temperature (with a gain of 2.5 for neutrons of $\lambda > 5 \text{ \AA}$) and a better pulse shape. However, it has to be recirculated in order to anneal out the radiation damage (in particular, atomic hydrogen).
- Shape of the moderator. This can be grooved for some instruments, like SANS, for which a sharp definition of the pulse is not important.
- A large unpoisoned and coupled moderator could feed several guides with instruments which require only coarse wavelength definition.

b. Monochromators

Single Crystals

Single crystals are widely used at reactor instruments and in inverse-geometry instruments at pulsed sources. In both cases, large-area analyzer crystals are needed. Development of better crystals is needed for the exclusive application of neutron research, because this has unique requirements for size and material used. These are:

- Growth of large crystals of beryllium, for high-energy neutrons.
- Growth of crystals with variable spacings ($\text{Si}_x\text{Ge}_{1-x}$) to increase the wavelength acceptance without increasing the mosaic spread.
- Manufacture of Ge crystals with anisotropic mosaic spread.
- Construction of doubly bent monochromators, eventually with d (plane spacing) gradient.

Multilayers

Currently, state-of-the-art thick multilayers have a d spacing of $\sim 50 \text{ \AA}$. With a d spacing of $30 \text{ \AA}$ or smaller, artificial multilayers may substitute mica with far superior diffracting power.

c. Neutron Guides

The classical neutron guide, a rectangular polished Ni-coated glass tube, transports neutrons over a large distance. With the glancing angle being $\gamma = \lambda \times 0.1^\circ/\text{\AA}$, the accepted solid angle increases proportionally to λ^2 . For that reason, nearly every reactor is today equipped with a cold source, feeding a bundle of guides. A $1/8$ sector at the reactor face accommodates some five guides and 10–15 instruments. In the U.S., there is only one guide hall (at NIST). If an upgrade of existing high-flux reactors is envisaged, a cold source *plus* guide hall and a selection of low-energy instruments is the obvious choice.

At a pulsed source, a well-coupled cold moderator will produce a high flux of long-wavelength neutrons with rather long pulses. Guides then feed instruments which do not use the time structure for resolution but rather increase intensity in a "multiplexing" mode. Examples:

- SANS.
- MUSICAL = backscattering, μeV resolution.
- NSE: TOF resolves Q , not energy.

The acceptance angle for *thermal guides* is rather small. For that reason, an instrument at a thermal guide is inferior to its analog at a beam port. This situation could change in the future, provided the necessary development is done. Supermirror coatings can increase the acceptance angle by a factor of 3 or even 4. At least the top and bottom mirrors should be treated in that way to increase the vertical divergence.

d. Focusing Devices

High-quality *imaging mirrors* need to be developed to improve the performance of a number of instruments, like NSE and SANS. An attempt is being made at NIST to use a series of cylindrically bent mirror segments in a polygonal arrangement to focus the SANS beam. However, the ratio of specular-to-diffuse reflected signal must be sufficiently high for satisfactory applications.

With *focusing monochromators*, the quasiparallel beam of a Ni guide can be focused both vertically and horizontally onto a small sample. A bent monochromator cut at 5 or 10° with respect to the reflecting plane, whose d spacing varies along the crystal, can even focus monochromatic neutrons in a rather wide energy range.

A tapering snout at the end of a guide focuses the beam at the expense of homogeneity.

More exotic devices which should also be considered are bundles of *capillary tubes* or microbender plates.

e. Choppers and Filters

High-speed *magnetic bearing* choppers serve for energy definition or background suppression. Further development of magnetic bearing technology should be pursued.

A fast *velocity selector* for SANS with $\Delta\lambda/\lambda = 10\%$ is now available. A radiation hardened version which could suppress higher order for triple-axis spectrometers and diffractometers at reactors still needs to be developed.

f. Polarizers

The use of polarized neutron scattering and polarization analysis offers a tremendous advantage for identifying the magnetic scattering and for separating coherent from spin-incoherent scattering. However, the efficiency of the available polarizers is low and needs to be significantly improved. *Supermirrors* provide a satisfactory solution for cold neutrons, in particular if used as transmission devices with transparent Si substrates. Polarizer crystals for thermal neutrons are of uneven quality and do not offer optimal reflectivity. Here, an intense research effort is necessary.

Improved cutoff-angle supermirrors would extend the useful wavelength range to the thermal region.

Polarized ^3He gas filters carry great promise. In principle, they represent the ideal solution for polarizing a white beam at the thermal and hot energies. Currently, several methods for optical pumping of ^3He are being studied, and a research effort should be concentrated on this subject. If this cannot be made to work, low-temperature filters, such as SmCo_5 , can prove to be interesting as analyzers; beam heating prevents their use as polarizers.

Neutron polarization is also the basis for several beam shaping tools like the Drabkin resonance monochromators or electronic choppers. These become viable only if the efficiency of the polarizers closely approaches 100%.

g. Detectors

In general, an increase of the area of a detector increases the efficiency of an instrument enormously. Thus, detectors are desired that cover as much as possible of the 4π diffraction space.

More intense neutron sources require detectors with shorter deadtimes. These are now, at best, 2 μs ; deadtimes below one microsecond are desirable.

For a conventional experiment, the spatial resolution required is no smaller than the size of the sample (1 mm for reflectometry; 1 cm for diffraction). However, tomographic applications might become of increasing importance. The detectors may be classified as follows:

Gas Detectors

In PDS's, the deadtimes are 5–10 μ s. The spatial resolution achieved in resistive wires is ~ 1 mm. In the new type of gas detectors (microstrip detectors using deposited wires with high-field gradient), the resolution is a few tenths of a millimeter.

Scintillators

The traditional scourge of these detectors, γ sensitivity, has been recently eliminated by a clever choice of material and electronic differentiation of the gamma and neutron signals. The best scintillator at present is ^6Li -loaded ZnS. Although this is rather opaque, its efficiency can be made practically unity for neutrons of wavelength 1 Å and longer. Most of the development took place at RAL, and its successful application required a lead time of ten years. The deadtime is ~ 2 μ s.

With special scintillators, channel plates, resolutions of the order of 10 μ m have been achieved. These have also been applied in neutron cameras.

Solid State

They are the least developed detectors for neutron applications. In all cases, special atoms absorb the neutrons and emit an α -particle. Various devices have been proposed:

- Bolometers: assemblies of superconducting Nb spheres. Local heating destroys the superconductivity of some spheres; the ensuing variation of magnetic field is recorded.
- Metal-insulator sandwich. The α -particle is measured by tunneling.

At present these devices have an efficiency no better than 1%.

4 Other Neutron Applications

4.1 Isotopes

C.W. Alexander (ORNL)

Y. Maruyama, M.D. (University of Kentucky), Chairman

R.C. Reba, M.D. (University of Chicago)

C. Sherlock (Chicago Bridge & Iron)

4.1.1 Update and Importance

Isotope usage and research has expanded tremendously, and there is extensive use of a variety of radioactive isotopes in medicine, agriculture, industry, and research at the present time. Daily medical use in hospitals and clinics is extensive. Neutrons have been one of the important nuclear particles which have brought about this progress. Some important achievements and contributions in the past decade include:

1. Isotope production for use in medicine, biology, agriculture, industry, geology, and research; notably transuranium isotopes. (This is the most successful example of technology transfer and collaboration in the DOE.)
2. Neutron treatment of radioresistant cancer using ^{252}Cf brachytherapy.
3. Nuclear medicine diagnosis, treatment, and research.
4. Industrial imaging and radiography with ^{192}Ir and ^{60}Co , and ^{252}Cf radiography of aircraft.
5. *In vivo* and *in vitro* activation analysis (e.g., thermal neutron airport luggage inspection for explosives).
6. Advances in heavy-element chemistry and physics and understanding of the fission process.

4.1.2 General Considerations

Hundreds of thousands of human lives are affected annually and thousands saved from deadly diseases, such as cancer, heart and metabolic disease, by radiation and nuclear medicine using radioactive isotopes. Recent beneficiaries included former President George and Mrs. Barbara Bush, who were successfully treated by ^{131}I for thyroid disease, which, in the case

of Mr. Bush, affected his heart. About 35000 medical isotope tests are performed daily. The quality, quantity, and availability of isotopes continue to be an important factor for progress and R&D in these fields.

Industrial radiography directly affects thousands of workers and produces estimated gross revenues of \$500M per year.

4.1.3 ^{252}Cf Neutron Brachytherapy of Cancer¹⁻⁸

A key advance has been the recognition of ^{252}Cf , which decays by alpha decay and spontaneous fission, as a means for neutron therapy of radioresistant cancers. Evidence from extensive basic cancer research studies has shown the high efficacy of neutrons in overcoming radioresistance to conventional radiation therapy. This has been shown for factors such as hypoxia, rapid tumor growth, necrosis in tumor, and the ability to recover from x-ray damage of types designated as sublethal or potentially lethal. Based on purely radiobiological rationale, neutrons should be highly effective in overcoming the radioresistance of cancers to conventional x-radiation therapy.

In the past three decades, there has been extensive study of neutrons for cancer therapy. This has focused on fast-neutron beam therapy where some efficacy has been found against the parotid gland, prostate gland, and sarcoma tumors, but with frequent side effects. The most successful form of neutron therapy for bulky tumors resistant to x-rays has utilized sealed sources of Cf implanted as seeds directly into the tumors. This form of therapy, termed "brachytherapy", delivers neutron radiation directly to the tumor and target volume and minimizes dose to adjacent normal and sensitive tissues. By this practice, brachytherapy has avoided the problem of normal tissue complications.

The methods for treatment are basically *intracavitary* (i.e., in body cavities like the vagina, anus, rectum, uterus, and cervix) or *interstitial* (i.e., inserted into tissues, e.g., prostate or mouth). For intracavitary therapy, sources about the size of a nail (25 mm $\times$ 2.8 mm) are available and effective. For interstitial therapy, tiny seed sources are needed and should measure about 5 mm $\times$ 1.5 mm in size and contain about 5 μg of ^{252}Cf . The sources should be aligned in plastic ribbons for clinical use. Seed and ribbon sources are able to conform to body tissues and tumors readily without discomfort, and they are highly versatile in use. ^{252}Cf is especially effective for treatment of cervix, uterus, vagina, rectum, anus, head and neck, and brain cancers. ^{252}Cf may be the only agent effective versus glioblastoma multiforme (brain cancer), where high efficacy has been demonstrated and survival times have been doubled. The cure rates for bulky, localized cervical cancers have been found to be 95% for 5 years and, even for advanced cases, can be 54% for 5 years. These observations have been confirmed in Japan, the former U.S.S.R., and Czechoslovakia.

Nearly 3000 cancer patients with tumors of a variety of sites have been treated to date with generally excellent results observed even in the earliest clinical trials. These early trials used primitive equipment and sources, and the trials were done with no prior knowledge on how to use neutron brachytherapy effectively and little or no understanding of radiobiology. In the future, neutrons can improve treatment of thousands of patients with otherwise untreatable cancers annually. Important technological and biological applications for future study include enhancement of the ^{252}Cf dose by boron and gadolinium thermal neutron capture during brachytherapy; design of a teletherapy machine for body surface or boron neutron capture therapy; ^{252}Cf research laboratory irradiators; and remote afterloaders and facility design for medical use. The present ^{252}Cf centers are projected to increase from 10 in 1990 to 100 in 2001, and each facility will require 500–1000 $\mu\text{g}/2.5$ years, and from 3 research irradiators to 10–20 irradiator/teletherapy machines by 2001. Each irradiator will require 25–100 mg every 2.5–5 years each.

Other cancer therapy uses have emerged using sealed sources, seed sources, alpha-emitting antibody/pharmacological chemicals, beta-emitting antibody/drugs, Auger electron therapy, and nonsealed agents for bone cancer therapy. A list follows:

Sealed Sources – ^{137}Cs , ^{192}Ir , ^{241}Am , ^{252}Cf (neutrons), $^{226}\text{Ra}^*$.

Seed Sources – ^{125}I , ^{192}Ir , ^{103}Pd , ^{145}Sm , ^{198}Au , $^{222}\text{Rn}^*$.

Bone Cancer Therapy – ^{89}Sr , ^{32}P , ^{153}Sm , ^{47}Sc .

Alpha-Emitter Therapy (Antibody/Pharmacological Agent/Receptor-Ligand) –

^{227}Ac , ^{211}At , ^{223}Rn , ^{212}Bi , ^{213}Bi .

Beta-Emitter Therapy (Antibody/Drug/Receptor-Ligand) –

^{186}Re , ^{188}Re , ^{67}Cu , ^{212}Bi , ^{213}Bi , ^{123}I .

Auger Electron Therapy – $^{195\text{m}}\text{Pt}$, ^{241}Am , ^{111}In .

^{89}Sr therapy for bone cancer represents an important recent advance for palliation of patients with breast or prostate cancer. Radioantibody therapy offers targeted therapy of cancer with fewer effects in surrounding normal tissues. One especially important product affecting daily human life is the smoke detectors containing ^{241}Am .

* Traditional source, now rarely used.

Table 4.1 lists important isotopes used for diagnosis, treatment, research, and development.⁹⁻¹⁵ The next generation ANS facility must assure the production of a continuous supply and availability of isotopes for biomedical uses and research and opportunities for advancing neutron science and applications in human medicine. In addition to production of these isotopes and research, a program for training and education of nuclear and radiochemists is important. Trained scientists are critical for the future of the field.

4.1.4 Industrial Radiography

Over the past thirty plus years, iridium 192 and cobalt 60 have been very widely used worldwide to perform industrial radiography. Recently, neutron radiography using ^{252}Cf has been used with good success. These first two isotopes are used to examine weldments on a variety of products such as nuclear submarines; repairs on nuclear system components, such as steam generators; petrochemical structures, such as hydrocrackers or reactors, process towers, and storage structures; penstocks and scroll cases for hydroelectric systems; wind tunnels; cryogenic storage structures; pipe lines; offshore platforms and moveable drilling rigs; high-vacuum facilities; basic oxygen furnaces; water storage structures and many others. Neutron radiography is used for detecting corrosion and debonding in military aircraft constructed of aluminum and/or composite materials. ^{192}Ir is used on steel or steel alloys with thicknesses through approximately 3 inches. ^{60}Co is used on steel or steel alloys with thicknesses through approximately 8 inches.

In the field, and in many manufacturing plant situations, ^{192}Ir is more economical to use than x-ray units. Likewise, in the field and in many shop situations, ^{60}Co is more economical to use in thicknesses over 3 inches than high-energy x-ray units of 2 MeV and higher. The reasons are versatility, duty cycle, and power requirements. The first is due to the smaller size and weight of the isotope exposure devices. This enables operators to maneuver the devices more easily for quicker set-up times between exposures. The second is because there is no duty cycle with isotopes as there is with x-ray units. The only limitation is the already shorter set-up time between exposures. The third is because isotopes do not need electrical power to operate.

All these briefly described favorable factors have resulted in the usage numbers for ^{192}Ir in the U.S. and Canada listed in Table 4.2.

Projected industrial radiography requirements for ^{192}Ir are expected to increase about 5% per year based on 1.38 million curies for 1992. ^{60}Co needs should remain at about 5000 curies per year.

Due to the decay of radioactive isotopes, it is essential to have a continual supply of radioactive ^{192}Ir and ^{60}Co for industrial radiography.

TABLE 4.1 Medical Isotopes Produced Via Neutron Reactions

Isotope	Use(s)
^{32}P	Treatment of cancer; cell metabolism and kinetics; labeled probes for molecular biology studies and human genome project.
^{33}P	"
^{35}S	Amino acid, protein, and molecular probe labeling.
^{46}Sc	Treatment of cancer.
^{47}Sc	"
^{51}Cr	Studies of blood cells; blood and bone marrow diseases.
^{55}Fe	"
^{59}Fe	"
^{60}Fe	"
^{64}Cu	Diagnostic studies of cancer and metabolic disorders.
^{67}Cu	Labeling monoclonal antibodies; cancer treatment.
^{75}Se	A convenient sulfur analog.
^{82}Br	Studies of metabolism; studies of estrogen receptor content.
^{89}Sr	Treatment of bone cancer.
$^{90}\text{Sr}/^{90}\text{Y}$	Treatment of painful and disabling arthritis; cancer treatment; beta plaque for eye.
$^{99}\text{Mo}/^{99\text{m}}\text{Tc}$	Diagnostic imaging.
$^{105}\text{Ru}/^{105}\text{Rh}$	Generator — radioantibody treatment of cancer.
^{103}Pd	Treatment of prostate cancer.
^{109}Pd	Treatment of painful and disabling arthritis; cancer treatment.
$^{109}\text{Cd}/^{109}\text{Ag}$	Generator — diagnostic imaging.
$^{117\text{m}}\text{Sn}$	Treatment of cancer.
$^{123\text{m}}\text{Te}$	Lung density measurements.
^{125}I	Radioactive seed therapy for prostate cancer.
^{131}I	Diagnose and treat thyroid disease and cancer; protein, peptide, and monoclonal antibody studies.
^{127}Xe	Cerebral perfusion/brain blood flow; pulmonary ventilation.
^{133}Xe	Pulmonary ventilation.
^{137}Cs	Brachytherapy of cancer.
^{141}Ce	Lung density measurements.
^{145}Sm	Radioantibody therapy.
^{153}Sm	Treatment of cancer.
^{153}Gd	Bone radiography.
^{159}Gd	Treatment of cancer.
^{165}Dy	Treatment of painful disabling arthritis.
^{166}Ho	" ; treatment of cancer.
^{169}Er	Treatment of painful disabling arthritis.
^{170}Tm	"
^{169}Yb	"
^{186}Re	Treatment of cancer.
^{188}Re	"
$^{191\text{m}}\text{Ir}/^{191}\text{Os}$	Generator — cardiac function, especially in infants and children.
^{192}Ir	Cancer treatment; industrial radiography.
^{194}Ir	Treatment of cancer.
$^{195\text{m}}\text{Pt}$	Diagnosis and treatment of cancer.
^{198}Au	Radioactive seed treatment of cancer.

TABLE 4.1 (Cont.)

Isotope	Use(s)
^{199}Au	Treatment of cancer.
$^{212}\text{Pb}/^{212}\text{Bi}$	Radioantibody treatment of cancer.
^{213}Bi	"
^{225}Ac - ^{229}Th	Cancer treatment research.
^{227}Ac	Radioantibody treatment of cancer.
^{241}Am	Low-energy brachytherapy of cancer.
^{252}Cf	Neutron brachytherapy of radioresistant cancers; irradiators.
^{253}Es	Monoclonal antibody labeling for treatment of cancer.
^{255}Fm	"

The list does not detail isotopes used in nuclear physics, chemistry, geology, industrial usage, environmental, or similar uses.

Without these isotopes for industrial radiography, the gap would have to be filled by the use of x-ray units and/or ultrasonic scanning. For ultrasonics to provide a permanent record such as produced by radiography is extremely costly. Also, ultrasonic testing is still less effective than desired for austenitic weld metal grain structures such as iron-chromium-iron or stainless steel. In either case, should funding for ANS not occur, the loss of a supply for isotopes when HFIR eventually becomes inoperable will lead to much higher industrial nondestructive testing costs.

In response to the Kohn Panel's request for further details on the current usage of isotopes, including transuranics, we have prepared tables providing additional information on current and projected isotope usage in the U.S. (Appendix B). Appendix B also includes tables of sales of reactor-produced isotopes for FY 1989-1991, an estimate of future U.S. demand for ^{89}Sr , and crude estimates of the current U.S. markets for ^{99}Mo , ^{125}I , ^{131}I , ^{133}Xe , and ^{32}P .

4.1.5 Proton-Rich Isotope Usage Needs

There are ~20 proton-rich isotope users in the U.S. and ~100 in the world. All of these facilities use medical cyclotrons for production, and since the isotope half-lives are short, the positron emission tomography (PET) — or other scanner — research is performed on-site. The present users have adequate supplies, and a spallation source will not affect user need or usage. BLIP or LANL may make a few proton-rich isotopes for on-site users, but this will not affect the general user community.

TABLE 4.2 Production and Use of Iridium 192
(numbers are approximate)

Year	Curies Produced (in Millions)	Shortage
1989	1.25	Negligible
1990	1.33	Negligible
1991	1.20	10%
1992	1.20	15%

4.1.6 Transuranium Isotope Production and Recovery

Transuranium isotope production and recovery and research on these isotopes was the primary reason for the construction and operation of the HFIR and the REDC at ORNL. Transuranium isotope production, recovery, and research continue as an essential and critically important program, which is vital to the mission of HFIR.

The HFIR-REDC complex produces and recovers the following mainline isotopes: ^{243}Am , ^{244}Cm , ^{249}Bk , ^{252}Cf , ^{253}Es , and ^{257}Fm (the heaviest isotope that can be made by neutron irradiation). In addition to the mainline isotopes, a number of derived isotopes are also produced: ^{248}Cm , ^{249}Cf , ^{254}gEs , and ^{255}Fm . With the exception of ^{252}Cf , these isotopes are exclusively used for research, and the HFIR-REDC complex is the sole producer of these isotopes in the western world. In terms of mission and scale, it is the only complex of its type in the world.

Research using these isotopes and elements is conducted in the following fields:

1. Nuclear reactions and synthesis of new species.
2. Nuclear properties of the transuranium elements.
3. Chemical properties in solution of the elements americium through fermium.
4. Spectroscopy of the heavy actinide elements.
5. Solid-state/magnetic properties of the transuranium elements.

6. Biomedical applications.

7. Industrial and other applications.

Research on transuranium elements was reviewed twice in the last decade by the National Academy of Science, and the research was determined to be important and essential.

Principal investigators and collaborators include all of the U.S. national laboratories, a number of U.S. universities and colleges, the ILL (France), GSI (Germany), Dubna (Russia), Karlsruhe (Germany), the PSI (Switzerland), JAERI (Japan), the University of Kassel (Germany), the University of Manchester (U.K.), Australian National University (Australia), the University of Leuven (Belgium), the University of Aarhus (Denmark), the University of Hanover (Germany), the University of Munich (Germany), Ruhr University (Germany), the University of Mainz (Germany), and others.

Two of the isotopes produced at the HFIR-REDC complex are reference production standards for the next-generation neutron source. These isotopes are ^{252}Cf and ^{254}gEs , and the amounts that the new source must be capable of producing on an annual basis are 1.5g of ^{252}Cf and ^{254}gEs .

The production of these two isotopes is complex both from a neutron and chemistry standpoint. The following neutron parameters are, generally, required to meet the production standard for ^{252}Cf and ^{254}gEs :

thermal neutron flux (2200 m/s flux): $\geq 1-2 \times 10^{15} \text{ n/cm}^2\cdot\text{s}.$

thermal-to-epithermal flux per unit lethargy ratio: $\geq 1:23.$

The high thermal neutron fluxes are required because of the large number of neutron captures required to produce ^{252}Cf and ^{254}gEs , the small cross sections of some of the nuclides (e.g., ^{246}Cm), the short half-lives of some of the nuclides (e.g., ^{253}Es), and the scarcity and value of the initial target material ($^{244-248}\text{Cm}$). The high epithermal fluxes are required to enhance the capture-to-fission ratios. This is extremely important to the production of ^{254}gEs which has a large thermal fission cross section. Production is also enhanced by an increase in the neutron temperature.

Coupled with these neutronic requirements is the requirement for heat removal. The primary mode of destruction or loss is neutron-induced fission. Fission cross sections for a number of nuclides are greater than 1000 barns. The heat removal requirement is highly dependent on the neutron spectra. However, the base heat removal requirement is on the order of 3-4 MW/m².

Downtime between cycles is also critical in determining the yields of the transuranium isotopes because of the short half-lives of a number of product nuclides. Quite often this is expressed as availability, capacity factor, fluence, or time-averaged flux. Downtime should be minimal in order to optimize yields and to reduce consumption of the valuable initial target material.

Fabrication and recovery of the isotope production targets is difficult and complex work and requires unique facilities. Because the transuranium elements emit gamma and x-rays, beta particles, alpha particles, and neutrons, the fabrication and recovery requires alpha-contained, remotely operable, and remotely maintained hot cells. Currently, the REDC is comprised of two adjacent hot cell facilities containing 16 hot cells and associated laboratories, and it is staffed for around-the-clock operation.

Over 20 years of experience in producing and recovering these isotopes has been gained at the HFIR-REDC complex. This experience and expertise has been translated into the conceptual design of the ANS. To our knowledge, virtually no experience exists for producing these isotopes in these quantities at a spallation neutron source. In addition, the requirement for high thermal neutron fluxes and high fluence or time-averaged flux exceeds the time-averaged fluxes for a state-of-the-art spallation source.¹⁵ A requirement that must also not be overlooked is the necessity of integrating the neutron source and the transuranium isotope production and recovery facilities.

Therefore, in regard to transuranium isotope production, experience and expertise favor production in the ANS. The ANS is also capable of providing the heat removal capacity required to cool the isotope production targets and the steady-state neutron fluxes and spectra essential to producing 1.5 g of ^{252}Cf and 40 μg of ^{254}gEs annually. In addition, the proposed site of the ANS integrates in the existing facilities at the REDC.

It appears much engineering would be required to produce transuranium isotopes at a spallation source.¹⁵ Roughly, an order of magnitude increase in time-averaged neutron flux will also be required to produce 1.5 g of ^{252}Cf and 40 μg of ^{254}gEs annually. If the spallation source were located at a site other than ORNL, the proper hot cell facilities will also have to be constructed.

4.1.7 General Isotope Production and Recovery

The production rate of isotopes can be simply defined as a function of the neutron flux, the energy-dependent production and destruction cross sections, and the decay constants of the nuclides. However, the production and recovery of isotopes consists of not only the above but, at a minimum, of the following parameters or requirements:

1. A range of neutron energies and fluxes.

2. Fluence, availability, capacity factor, or time-averaged flux.
3. Irradiation volume.
4. Heat removal.
5. Shielding.
6. Isotope target accessibility.
7. Associated facilities for the fabrication of isotope targets and the recovery of the isotope products.

Currently, isotopes are produced in reactors with thermal neutron fluxes in the range of $\sim 5 \times 10^{13}$ to 2×10^{15} n/cm².s and with thermal-to-epithermal flux ratios of ~ 5 to 40. Fast neutron fluxes (greater than 0.1 MeV) range from 0 to $\sim 10^{15}$ n/cm².s. These ranges are essential for the production of the widest possible variety of isotopes.

No one neutron source (steady-state or pulsed) can be built to optimize the production of all isotopes of interest. Therefore, as a first choice, the source that can provide the greatest range and greatest flexibility should be selected. Relative to isotope production, the ANS appears to be the first choice. However, a spallation source has capabilities that a fission-based steady-state source cannot provide. The most important capability is that neutrons are generated at energies in excess of the mean fission energy of ~ 2 MeV. This allows for the production or increased production of isotopes produced by threshold reactions such as (n,p) and (n,d). Many of these isotope products are of interest to the medical community because they can be produced carrier- or near carrier-free. In addition, it may open up additional research opportunities into neutron-rich isotopes.

4.1.8 References

1. Y. Maruyama, *Nucl. Sci. Appl.* **1** (1989), 677-748.
2. Y. Maruyama, J.L. Beach, and J.M. Feola, *Nucl. Sci. Appl.* **2** (1986), 187-826.
3. H. Yamashita *et al.*, *Nucl. Sci. Appl.* **2** (1986), 453-466.
4. A. Tsuya *et al.*, *Nippon-Acta Radiol.* **39** (1979), 389.

5. K. Kaneta *et al.*, in *Treatment of Radioresistant Cancers*, M. Abe *et al.*, eds. (Elsevier, North-Holland, Amsterdam, 1979), 97-114.
6. B.M. Vtyurin, *Review of Information Medical Public Health Services Oncology* (Moscow, 1981), 1-150.
7. T. Tacev *et al.*, *Nucl. Sci. Appl.* **4A** (1991), 201-212.
8. J.G. Wierzbicki, , Y. Maruyama, and C.W. Alexander, *Nucl. Sci. Appl.* **4A** (1991), 361-366.
9. *Separated Isotopes: Vital Tools for Sciences and Medicine*, Report of a National Research Council Workshop (National Academy Press, 1982).
10. *Review of the OHER Research Program on Nuclear Medicine*, Report of the HERAC, OER, Department of Energy (1989).
11. *Training Requirements for Chemistry in Nuclear Medicine, Nuclear Industry and Related Areas*, Report of a National Research Council Workshop (National Academy Press, 1988).
12. *The Role of a High-Current Accelerator in the Future of Nuclear Medicine*, Proceedings of a LANL Workshop, LANL Report LA-11579-C (1989).
13. *A National Biomedical Tracer Facility: Planning and Feasibility Study for the DOE* (Nuclear Medicine Press, 1991).
14. D.E. Erb, *et al.*, *A National Biomedical Tracer Facility*, presented at the Am. Chamber Soc. 1990. Transuranium Isotope Production Reference.
15. G.H. Lander and V.J. Emery, *Nucl. Instrum. Methods B* **12** (1985), 525-561.

4.2 Materials Irradiation

R.C. Birtcher (ANL)
A.N. Goland (BNL), Chairman
R. Lott (Westinghouse Science and Technology Center)
G.R. Odette (University of California – Santa Barbara)

The future success of the nuclear power option (fission and fusion) in the U.S. depends critically upon the continued existence of a healthy national materials-irradiation program. Consideration of the requirements for acceptable materials-irradiation systems in a new neutron source has led the working group to identify an advanced steady-state reactor (ANS) as a better choice than a spallation neutron source. However, the working group also hastens to point out that the ANS cannot stand alone as the nation's sole high-flux mixed-spectrum neutron irradiation source in the next century. It must be incorporated in a broader program that includes other currently existing neutron irradiation facilities. Upgrading and continuing support for these facilities must be planned. In particular, serious consideration should be given to converting the HFIR into a dedicated materials test reactor, and long-term support for several university reactors should be established.

4.2.1 Introduction

Our objectives in this report are to: (1) describe the broad range of neutron irradiation facilities required to support the DOE's mission to sustain and develop the nuclear energy option and (2) address specifically the potential roles of high-flux mixed-spectrum reactor and spallation-based neutron sources.

Understanding and optimizing the behavior of materials in neutron radiation environments is a pre-eminent concern to all nuclear energy technologies ranging from the current generation of fission to future fusion reactors. Ultimately, the safety, economics, and, indeed, the basic viability of nuclear energy systems depend on reliable materials performance. The effects of neutron radiation are manifested in diverse, profound, and, most often, deleterious ways: gross swelling and other dimensional instabilities; severe reductions in ductility, fracture resistance, and creep strength; and increased susceptibility to chemical attack. Significant irradiation damage is experienced by essentially all materials over a wide range of neutron fluxes, fluences, temperatures, and stresses.

4.2.2 Response of Materials to Neutron Irradiation

Property changes are controlled by the *combination* of a large number of environmental and material variables. Hence, predicting and improving the performance of materials in irradiation environments is far from a matter of "testing"; rather, success in meeting these objectives must proceed from a solid, science-based understanding of the fundamental underlying processes. Although they are manifested in different ways, these fundamental processes are generic to all forms of irradiation damage and include:

- Production of point defects and small defect aggregates in displacement cascades and transmutants constituting the *primary source of damage*.
- *Microstructural and microchemical evolution* resulting from long-range diffusion and interactions of the mobile primary defects in the form of various extended defect clusters, regions of nonequilibrium solute segregation, enhanced and induced precipitation, and modification of the pre-existing microstructures and precipitate phases.
- Microstructurally and microchemically sensitive physical, chemical, and mechanical *properties changes*.

The long-term objective of radiation damage research is to evaluate these fundamental mechanisms based on combinations of theory and experiment and to incorporate them into quantitative, predictive models of irradiation damage. Materials-irradiation facilities are needed to develop an understanding of the underlying mechanisms and to validate and calibrate the integrated materials performance models. It might also be noted here, that this fundamental research on defect physics, microstructural and microchemical evolution, and the micromechanics of deformation and fracture has impacted, and will continue to impact, the broad field of materials science.

4.2.3 Scientific and Technological Needs and Opportunities

This section is organized as follows: we begin by briefly outlining some of the significant technological motivations for irradiation damage research; we next provide examples of some key scientific opportunities; and finally, we describe the irradiation facilities needed to support this research endeavor.

4.2.3.1 Technological Motivations

- Ensure the continued safe and reliable operation of existing light water reactors (LWR's) including, when possible, extending their operating life.
- Support the development of advanced thermal, fast breeder, space, and defense fission reactors.
- Support the development of fusion reactors with particular attention to low activation materials.
- Support the development of accelerator-based neutron sources for tritium production, waste transmutation, and scientific applications.

4.2.3.2 Scientific Needs and Opportunities

It is necessary for the nation to maintain a group of neutron sources that provides the diverse neutron spectra required for different reactor environments, such as LWR, breeder, or fusion reactors. While an understanding of effects peculiar to a given technology are required, it is also necessary to maintain a strong research program on the fundamental processes described in Section 4.2.2. Such a general information base will allow connection of results to various applications.

The next generation of neutron sources should provide a range of neutron fluxes and controlled irradiation environments so that it is possible to address and resolve the following radiation-effects phenomena and questions:

1. A detailed description of the number and configuration of defects generated by the primary displacement cascade. These parameters include both stable defect complexes and freely migrating defects and should be determined as functions of neutron spectrum, neutron flux, neutron fluence (time-integrated flux), temperature, material type, and existing defect microstructure. The stability of defect structures should be determined as a function of time after their production for different temperatures and microstructure states.
2. Evolution of defect-induced microstructure and microchemistry is a multi-parameter problem. The interaction of migrating defects and transmutation products with each other and with a developing microstructure should be

determined. It is important that modeling and experiment interact strongly to allow extrapolation to new situations.

3. Investigation of nonequilibrium phenomena induced by radiation damage such as metastable phase formation, segregation, and amorphization.
4. Investigation of the role of electronic excitation in defect production and microstructure and microchemistry evolution in nonmetals.
5. Understanding of dose-rate effects in microstructure and microchemistry evolution.
6. Connection of property changes to microstructure and microchemistry evolution with the aid of detailed modeling.
7. Interaction between a material and its surrounding chemical environment.
8. Material property changes caused by irradiation-induced alterations in internal interfaces.

4.2.4 Existing Facilities for Controlled Neutron Irradiations and Potentials for Upgrade of Facilities

4.2.4.1 Existing Facilities

The description of radiation damage is the study of the effects of radiation on the structure of materials. This review report is concerned only with neutron radiation damage. With the broad definition of the discipline, any reasonably intense source of neutrons represents a potential resource. This review process was prompted by the need to evaluate two particular types of neutron sources, the ANS, and a spallation neutron source. To complete this evaluation, it is important to understand how these sources fit into the array of currently available neutron irradiation facilities. A brief summary of neutron irradiation facilities currently available in the U.S. has been compiled to provide perspective for these discussions. Historically, both nuclear reactors and accelerator-based charged-particle sources have been employed in the study of irradiation damage.

4.2.4.1.1 Reactor-Based Facilities

The vast majority of neutron irradiation damage studies have been based on nuclear reactor irradiations. The working group believes that this trend will continue through the foreseeable future. Nuclear reactors offer several inherent advantages:

- They allow large irradiation volumes.
- They have the potential for reaching high neutron fluences.
- Their radiation environment is generally well characterized.

Different reactor designs offer a variety of irradiation environments that can be characterized in terms of their neutron spectrum and the maximum available fast flux (generally defined as neutron energies greater than 0.1 MeV). *No single reactor irradiation facility can meet all of the needs of the radiation damage community.* Comparisons of damage states produced in various reactor spectra are required to study the processes of radiation damage. Radiation damage poses many significant technological concerns for existing LWR's and advanced reactor designs (including fast breeder and fusion reactors). The study of these problems requires neutron environments that simulate actual application environments. These considerations help to define the need for reactor irradiation facilities.

A summary of reactors currently available for controlled materials irradiations is included in Table 4.3. For the purposes of this discussion, these reactors have been divided into four basic categories. University research reactors provide a valuable resource for the study of radiation damage in LWR pressure vessels. (The working group is aware of four reactors that could be used for this purpose.) Higher flux test reactors, such as the ATR, have provided important sources of information for defense-related reactors and have significant potential for the simulation of LWR core environments. HFIR at ORNL has provided a rich source of data on materials with high helium-to-displacement-per-atom (dpa) ratios, which has been useful in the fusion program. HFIR irradiations have also aided in the understanding of material behavior for LWR applications. The breeder reactor test facilities (EBR-II and FFTF) are required for breeder reactor alloy development programs.

4.2.4.2 Accelerator-Based Neutron Sources

Spallation neutron sources produce neutrons as a result of the destruction of nuclei within a heavy metal target by high-energy protons. The neutron generation increases with increasing proton energy, and beams with energies over 450 MeV are typically used with the beam delivered in a pulsed mode. The neutrons are produced with the same time structure as the incident protons.

TABLE 4.3 Summary of Reactors Currently Available in the U.S. for Controlled Materials Irradiations

Type	Spectrum	Peak Fast Flux [E > 0.1 MeV] (n/cm ² ·s)	Technologies Supported
University Research (4 possible)	Mixed	< 10 ¹³	LWR pressure vessel
Test Reactor (ATR) (Nat'l Labs, e.g., HFBR)	Mixed	> 10 ¹⁴	Defense, potential for LWR core
High Flux (HFIR)	Mixed	> 10 ¹⁵	Fusion, LWR
Fast Breeder (FFTF, EBR-II)	Fast	> 10 ¹⁵	Fast breeder

Such neutron sources are characterized by neutron spectra resembling a degraded fission spectrum with an additional component of very high energy neutrons. These high energies extend up to the incident proton energy. Neutrons are generated within a fairly small volume determined by proton stopping so that the resulting fluxes have strong spatial gradients. Two spallation neutron sources are operating in the U.S.

ANL/IPNS: In its original configuration, the IPNS had separate targets dedicated to radiation effects and neutron scattering. The Radiation Effects Facility was closed in 1984. Originally, the IPNS Neutron Scattering Facility had a depleted uranium target. This was replaced with highly enriched uranium resulting in a factor of 2.5 increase in neutron generation. Protons are delivered to the target in 100 ns pulses at variable repetition rates up to 30 Hz and time-averaged currents of 14 to 16 μ A. The dedicated Radiation Effects Facility at IPNS had a time-averaged fast neutron flux of 2.8×10^{12} n/cm²·s for 14 μ A of 450-MeV protons. With an enriched uranium target, the flux would be 7×10^{12} n/cm²·s.

The irradiation thimble currently in use within the Neutron Scattering Target Assembly is not in an optimized position and has a fast neutron flux of 2×10^{11} n/cm²·s with the depleted uranium target. An enriched uranium target would increase the flux to 5×10^{11} n/cm²·s.

LANL/LAMPF, LASREF: The LASREF is located at the beam stop of LAMPF. It operates with a time-averaged proton beam of 800 μ A at 800 MeV. Neutron generation at LASREF depends on the composition of isotope production target and proton irradiation experiments located in front of the copper beam stop. The time-averaged fast neutron flux is about 3×10^{13} n/cm²·s for 1 mA of 800 MeV protons.

4.2.4.3 Potential Upgrade of Existing Facilities

4.2.4.3.1 Reactors

The working group is extremely concerned about the erosion in the number of reactor irradiation facilities. We believe that the reactors listed in Table 4.3 represent a minimum acceptable level of research activity. In the near term, we recommend that all of these reactors be maintained as irradiation facilities. Further development of the irradiation facilities at these reactors would provide the foundation for a comprehensive radiation-damage program.

4.2.4.3.2 Accelerator-Based Sources

A proposed upgrade of IPNS would utilize a proton energy of 1500 MeV and a proton current of 500 μA . This would result in an estimated fast neutron flux of $2.5 \times 10^{14} \text{ n/cm}^2\cdot\text{s}$ in an optimized facility used by radiation effects and neutron scattering.

4.2.5 Strengths, Weaknesses, and Complementarity of Reactor Spallation vs Neutron Sources

In order to address the relative merits of proposed new sources with respect to their ability to support meaningful materials-irradiation facilities, it is necessary first to define the desired characteristics of such facilities. In general, they can be separated into two classes: those that establish the quality of the facility, and those that are imposed upon the facility by special requirements associated with specific technological needs.

4.2.5.1 Characteristics that Define Facility Quality

The first category consists of those attributes that any dedicated, modern materials-irradiation facility must possess. Among the principle entries on this list, one stands alone as the single most important: the facility must provide a capability to perform controlled, single-variable experiments. An irradiation space that merely permits high levels of sample exposure to neutrons under conditions of uncontrolled neutron flux, spectrum, and experimentally relevant variables is

unacceptable. This imposes upon the facility a set of characteristics/requirements that generically define its quality:

1. First and foremost, the radiation environment experienced by the sample(s) must be well characterized by an adequate, ongoing, documented dosimetry program.
2. Control of the experimental parameters implies a capability to perform single-variable experiments. Temperature measurement and control represent the most important of these capabilities; in broader terms, the facility must be able to support *in situ*, real-time electrical measurements of properties such as electrical resistivity and others that also generate electrical signals (see item 6 below).
3. Realistic consideration of the scope of a meaningful irradiation effects program has established the necessity for a large experimental volume in the appropriate-flux region of the source. A typical volume would be 5–20 L.
4. Access to sample capsules for removal and replacement at arbitrary times would expand the range of feasible experiments.
5. Flux gradients over the sample volume must be minimized in order to limit complications encountered in the subsequent interpretation of the experimental data. Gradients that exceed prescribed values or fluctuate grossly with time are unacceptable.
6. As temperature control is always required, excessive nuclear (gamma) heating cannot be accommodated in an experimental plan. As a general stipulation, the lower the gamma heating rate, the better, although some gamma heating can be helpful.
7. Many of the required irradiations will be of long duration. Interpretation of the data will be simplified if the neutron spectrum remains constant during the radiation exposure. It is difficult to conceive of a realistic program for monitoring spectrum changes that occur frequently. Even those that are documented on a periodic basis over two to three years would introduce serious difficulties in data analysis and could jeopardize the objectives of the experiments.

4.2.5.2 Characteristics Required by Specific Technology Questions

The second category of facility characteristics consists of those most closely associated with special technological demands. While such a list cannot be exhaustive, it is likely to include at a minimum:

1. The necessity to reach significant total exposures (fluences) in reasonable times implies a high rate of atomic displacements. The fusion and breeder reactor technologies each require this feature. Significant microstructural and microchemical changes do not develop or are undetectable at low fluences.
2. Fusion and accelerator materials development requires irradiations that ensure the appropriate high ratios of helium-to-dpa. These ratios must be attainable in the facility.
3. For reasons related once again to the fusion program, it must be possible to supply high-energy neutrons (14 MeV). This capability would span the range of interest that the materials-irradiation community is likely to require for the foreseeable future.
4. An ability to match required spectra including methods for spectral tailoring has become a desirable feature for future experiments. It enhances the utility of a given source and permits the pursuit of certain experiments that otherwise could not be conducted.
5. Recent experience with LWR technology has led to the realization that a relatively low-dose-rate facility is needed in addition to facilities already mentioned.
6. Certain components in fusion reactors will be exposed to neutrons while operating at cryogenic temperatures, viz., superconducting magnets. Proper investigation of the consequences of these exposure conditions requires a cryogenic-temperature irradiation facility of modest flux. Such a facility also would serve as a welcome adjunct to a program in basic radiation effects whereby primary radiation-induced defects can be immobilized and subjected to subsequent thermal annealing studies.

4.2.5.3 Summary

This analysis of the characteristics of an "ideal" materials-irradiation facility made it possible to evaluate each type of source, recognizing that some present design uncertainties could not be resolved.

It should be apparent that no single neutron source will be capable of meeting all of the foregoing criteria. Thus, it is already clear that the only viable solution to a national strategy for maintaining a healthy materials-irradiation program is one that embodies more than one source. It is stated here, and will be reiterated later, that it is in the national interest to maintain at least some of the remaining irradiation capabilities still available at universities, industrial sites, and national laboratories.

4.2.6 Conclusions and Recommendations

4.2.6.1 Conclusions

The conclusions drawn by this working group are based in part upon operating experiences drawn from existing reactors and spallation neutron sources and upon the characteristics of the proposed ANS and of a spallation source capable of providing a 1–2-MW proton beam on target. A detailed design of the spallation source was not provided; the assumption was made that the proton energy would lie between 800 and 1600 MeV and that the neutron yield increases linearly with proton energy.

In addition to the assumptions of a technical nature outlined above, the working group also assumed that the U.S. will continue to depend upon nuclear power systems far into the next century. Nuclear power, for the purposes of this discussion, includes fission and fusion technology and applies to applications in space and for defense purposes, as well as for meeting conventional civilian power demands.

Advanced concepts associated with the fission and fusion power options will require a broad materials-irradiation program that must be based upon the availability of diverse neutron sources. It is clear that the ANS, whose primary purpose is to support research with thermal neutron beams, will not suffice as the sole high-flux source for national materials-irradiation program needs. Table 4.3 of this report lists currently available options for additional neutron sources in this country. Some of these sources are scheduled for shutdown or are already shutdown in a standby mode.

Finally, these facts together with those stated in Section 4.2.5 have led to several conclusions:

1. The relatively low time-averaged fast flux and limited irradiation volume of a spallation neutron source are major negative factors.
2. Conclusion 1 leads to the choice of a steady-state reactor option, such as the proposed ANS, but with reservations.
3. Reservations concerning the ANS can be addressed by further design considerations in order to establish more firmly its utility to a comprehensive materials-irradiation program. These design considerations would affect core and reflector regions of the reactor.
4. Regardless of the final configurations of ANS irradiation facilities, it should be reiterated that the ANS will not be adequate as the sole future materials-irradiation facility for the nation.
5. Although a higher priority is assigned to the construction of the ANS, materials-irradiation experiments of limited scope could be performed in a high-flux spallation neutron source.

4.2.6.2 Recommendations

1. Move ahead with the construction of the ANS, taking additional time to optimize its design and operating parameters as best as possible for the many research communities it will serve.
2. Develop a strategy for maintaining or, if possible, upgrading some number of the nation's existing irradiation facilities (see Table 4.3) for long-term use.
3. Consider funding a design study for converting the HFIR into a dedicated materials test reactor. This would involve replacement of its core but not its pressure vessel. A rough estimate of the cost of this conversion is \$150M (FY 1992 dollars).
4. Include plans for materials irradiation facilities in any strategy for constructing a new spallation neutron source.

4.3 Materials Analysis and Imaging

R.F. Fleming (University of Michigan), Chairman

T.Z. Hossain (Cornell University)

R.M. Lindstrom (NIST)

W.J. Richards (McClellan Air Force Base)

The use of proper neutron reactions (as opposed to neutron scattering) for analytical purposes and of radiography for imaging has been widespread for decades, with some of the most fruitful methods and applications having been developed at small reactors (1 MW and below) around the world. These methods (neutron activation analysis, prompt-gamma activation analysis, neutron depth profiling, and neutron radiography) add only a small incremental cost to the operation of a neutron source, but deliver useful and visible results and serve as a major training ground for nuclear chemists and radiographers. Because of their relative freedom from chemical effects, their well-understood systematic errors, and stability over long periods of time, nuclear methods of analysis play an important role in calibrating reference materials and analytical sensors used to maintain statistical quality for industrial process control. Most of the in-core irradiation applications are not limited by the available flux density: it is usually easy to produce sufficient radioactivity by irradiating a large enough sample for a long enough time. Similarly, neutron radiography imaging, both film and real time, is being accomplished satisfactorily at present with acceptable exposure times. The beam techniques of analysis are intensity limited, so that high-intensity beams, especially cold beams, are needed.

As the number of research reactors shrinks, the neutron analysis community will need to adapt to fewer large sources, so these facilities must plan to accommodate visiting researchers in these fields to a greater extent than in the past.

4.3.1 The Scientific Case

Neutron-based analytical methods in the past decade have for the most part changed in an evolutionary rather than revolutionary way. They have continued to be a powerful tool in the analytical kit, despite the wealth of other methods that have become available from the manufacturers of analytical instruments.

Because of its excellent sensitivity for measuring very small quantities of elements nondestructively, without concern for contamination during sample dissolution, neutron activation analysis remains essential for the analysis of high-purity natural or man-made materials such as human body fluids and semiconductor silicon. Neutron activation has shown in the past decade that the role of several toxic and essential trace elements in human metabolism has been misunderstood because of the inability of other analytical techniques to measure the very low levels

in normal body fluids with certainty. Only neutron activation can measure with sufficient accuracy and sensitivity the low levels of contaminants necessary to assure good yields of multimegabit memories and other very large-scale integrational (VLSI) circuits. For the manufacturing of dynamic random access memory (DRAM) chips, it is essential to screen materials for uranium and thorium contents. The natural alpha radioactivity from these two elements induces undesirable "soft-error" effects, disqualifying them for use. Materials for the current generation of DRAMS (16 megabit) can barely be screened for U and Th with the neutron fluxes available. For the next generation (64 to 256 megabit), neutron activation with higher flux will be critical for developing the necessary high-purity materials. The essential role of extraterrestrial impacts in geological mass extinctions was detected and verified by neutron activation, and this remains the only method of measuring the iridium anomalies in large numbers of geological samples. The bulk of multielement analysis of large numbers of samples in fields as diverse as geology, archaeometry, and air pollution chemistry is performed by neutron activation.

Neutron beams have come into routine application to analytical problems in the past decade as prompt-gamma activation analysis and neutron depth profiling have matured. The increasing sophistication of beam filters and cold source design, coupled with tremendous advances in imaging technology, now allow neutron radiography to provide material inspection information previously unavailable. A specific example is the fact that real-time neutron radiography has become a standard tool in studies of two-phase fluid flow, lubricant flow in engines, and detection of low-level corrosion in aircraft structures.

4.3.2 Reactors and Spallation Sources

For measurements exploiting proper nuclear reactions the following criteria are important in selecting a neutron source.

For irradiations in neutron fields (i.e., neutron activation analysis):

1. Availability: neutron hours per year.
2. Volume in which the flux exists and into which the sample may be inserted.
3. Magnitude of neutron flux on the sample: neutrons per square centimeter per week.
4. Radiation heating and neutrons of undesirable energies.

5. A pure, tailored spectrum for each of the following:

Thermal: most important.

Epithermal: 0.1 eV to tens of electron volts, to suppress activation of major interferences.

Fast: greater than 100 keV, for (n,X) reactions (e.g., ^{39}Ar - ^{40}Ar dating).

For irradiations with neutron beams:

1. Availability: neutron hours per year.
2. Pure, cold beams with no fast neutrons or gamma rays are best because reaction rates go as $1/v$; intense epithermal beams are useful for resonance neutron radiography.
3. For radiography, both large-area and high L/D beams for high resolution are required. For reaction techniques, both large-area (for large samples) and small spot beams are needed; the latter may be focused for greater intensity.

If these criteria could be met, spallation sources and reactors would be equivalent. But for elemental analysis there must be a *unique target* nucleus for each radioactive species created, so therefore, there must be no neutrons above 20 MeV. Even 14 MeV neutrons are a problem in this regard. For most applications, a pulsed source is undesirable; in rare cases, a time mark might be exploited. It is our understanding that the epithermal and fast components of a spallation-source spectrum are too high for most neutron activation analysis, and that the instantaneous neutron and gamma rate at the pulse time is too high for beam techniques. Unless pulsed neutrons could be fed to a quasi-steady-state cold source, we conclude that, for materials analysis and imaging, a spallation neutron source would need to be supplemented by regional research reactors with cold sources.

4.3.3 New Opportunities for Tenfold Higher Flux on Sample

The most dramatic improvements from a tenfold increase in flux would be seen in the beam techniques, where the counting rate is currently low, rather than in-core irradiations. Neutron tomography would be greatly improved in resolution and throughput, with practical consequences for its use as a production technique. For neutron depth profiling, such nuclides as ^{17}O and ^{14}N would be measurable in hours rather than days, so that systematic multisample studies of oxide and nitride systems could be performed in a reasonable time. With a tenfold higher event rate in beam techniques, coincidence counting rates would become analytically practical, with great improvement in the specificity of analysis. In conventional neutron activation, a tenfold increase in flux density would immediately translate into a tenfold smaller analytical sample size for most materials, or a higher throughput, assuming that the detection efficiency of the induced radiation is unchanged. In some cases (notably semiconductor silicon), where the detection limit is determined

from the signal strength rather than the signal-to-noise ratio, a tenfold lower detection limit in concentration may be anticipated. The use of narrow-bandpass filtered beams for cross-section measurements and nuclear structure studies has always been limited by low intensity: tenfold higher source strength would expand the opportunities available. For higher resolution radiography ($L/D > 500$), more intense beams would be advantageous.

4.3.4 Short-Term Improvements with New Instrumentation

Reflective optics are beginning to be developed for focusing neutron beams onto a small spot, which offers the prospect of higher flux on target and compositional imaging with prompt radiation. Sophisticated coincidence counting instrumentation and techniques have been developed in nuclear physics which need to be adapted to the needs of analysts with regard to data rate and ease of use.

Because the energy of cold neutrons is below the Bragg cutoff energy of most metals, cold-neutron radiography gives greatly improved contrast for otherwise impenetrable thick samples. No cold beams are available in this country for radiographic techniques to be investigated; one should be developed either as a dedicated facility or in conjunction with existing cold-neutron instruments. New imaging technology — charge-coupled devices (CCD's), charge-injected devices (CID's), and storage phosphors — promise faster, higher resolution radiographs.

4.3.5 Recommendation

If a CW neutron source (e.g., the ANS) is constructed, then we suggest that a family of research reactors with cold sources be established on university campuses to develop the next generation of scientists, instrumentation, and measurement techniques. If a spallation source is chosen, these reactors would be *required* in order to continue productive work in materials analysis and imaging.

4.4 Fundamental and Nuclear Physics

G.L. Greene (NIST), Chairman
J.M. Pendlebury (ILL, France)
S.J. Seestrom (LANL)
S.A. Werner (University of Missouri - Columbia)

4.4.1 Introduction

The development of intense neutron sources for neutron scattering and materials science research has engendered a vigorous program of research in which the neutron, rather than serving as a probe, is itself the object of study. The scientific content of this program has relevance for particle physics, nuclear physics, and astrophysics, as well as for tests of fundamental phenomenology, for the determination of fundamental constants, and for the investigations of the fundamental symmetries of nature. This work, with its extremely eclectic collection of activities, has come to be referred to as *fundamental neutron physics*. Along with more traditional investigations in nuclear physics, this area of activity has flourished with access to the high neutron beam intensities at modern high-flux reactors. In addition, this work has benefited from the availability of intense *cold* neutron beams and from the development of neutron optical techniques which include highly efficient neutron polarizers and perfect single-crystal interferometers.

Most, if not all of these activities will benefit substantially from higher neutron fluxes. A significant fraction of the experiments in fundamental neutron physics are currently strictly count-rate limited, and in many cases, an order of magnitude increase in flux would result in a qualitative change in the investigations which may be envisaged. That is, with substantial improvements in flux, there are interesting *feasible* measurements which would not otherwise be attempted.

As an aid in the review of this area of research, we have divided the research which comprises "fundamental" and nuclear physics research with neutrons into four categories. The first, *fundamental and particle physics with cold neutron beams*, involves neutron beam research and is concerned with elementary particle physics, tests of fundamental symmetries, and the determination of fundamental constants. *Particle physics with stored ultracold neutrons*, concerns research with neutrons of sufficiently low energy that they can be "trapped", held, and studied in material bottles for long periods (i.e., minutes). *Neutron interferometry* is concerned with a class of experiments in which the wavelike character of neutron beams is exploited using techniques which are analogous to the interferometric methods familiar from classical optics. *Nuclear physics* concerns efforts in which the neutron is used as a probe in the investigation of the structure and interactions within the nucleus. We note that this classification is not rigorously distinct, with a number of activities spanning two or more categories. In that sense, our classification is somewhat arbitrary. This overlap is in fact an important feature of this area of research, for the field of fundamental neutron physics is actually a rather broad interdisciplinary endeavor. The "cross-fertilization" between different areas of physics has been essential to the vitality of this research.

4.4.2 Fundamental and Particle Physics with Cold Neutron Beams

There are, in general, two reasons why cold beams are well suited to the study of the neutron and its fundamental interactions. Experiments which measure static neutron properties (including decay properties) or the interaction of free neutrons with laboratory fields typically benefit from the long observational TOF's which are available with low-energy neutrons. Experiments which require spin-polarized neutrons profit from the relative ease with which intense cold beams can be polarized with very high efficiency. Fundamental research with cold neutron beams includes a rather considerable variety, and it is quite beyond the scope of this report to provide a complete review. Aside from focusing on a few specific (and perhaps arbitrary) highlights, this section will present only the briefest survey of the field. More detail is readily available in the form of the proceedings of a series of international workshops and conferences¹⁻⁴ devoted to this work.

Accurate values for the properties of the free neutron which have been determined using cold neutron beams have implications in a variety of fields.* The determination of the neutron magnetic moment can shed light on the static quark structure of the nucleus. Limits on the neutrality of the neutron reflect on the more general question of the neutrality of matter and indirectly on the gauge invariance of the electromagnetic interaction. Measurements of the correlations in free neutron decay can shed light on the origin of time-reversal symmetry violation, as well as providing important information on the nature of semileptonic weak interactions. Measurement of the neutron beta-decay lifetime is also required for detailed tests of the standard model of the weak interaction, as well as providing a parameter of great importance in astrophysics and cosmology. The measurement of selected neutron cross sections is important for stellar astrophysics and for the understanding of the details of the standard solar model. The accurate determination of the interesting quantity given by the ratio of Planck's constant to the neutron mass (h/m_n) can provide an important input to the least-squares adjustment of the fundamental constants, as well as providing an independent determination of the fine structure constant α . Measurements involving polarized neutrons have been used to shed light on the details of the weak interaction between quarks. A particularly interesting program of research carried out over the last decade concerns the search for a baryon non-conserving "oscillation" (in parameter space) between the free neutron and its antiparticle, the antineutron.

It has long been realized that among the quantum numbers which characterize the neutron and antineutron (i.e., charge, spin, etc.), only the value of baryon number, B , differs between the two. Baryon number has long been thought to be an exactly conserved quantity, though it should be noted that there is no compelling theoretical principle which requires this conservation. As the neutron and antineutron differ only by baryon number ($\Delta B = 2$), it can be argued that (insofar as we understand) only baryon number conservation prevents the spontaneous transition (or

* Except where specifically appropriate, we will dispense with references to specific research papers and restrict ourselves to the citation of reviews and conference proceedings in which more complete bibliographies may be found.

oscillation) between the neutron and the antineutron. Such particle-antiparticle oscillations are known to occur, most notably in the K^0 meson system.

With the development of super-symmetric, grand unified theories (GUTS), the possibility that baryon number was not exactly conserved became a crucial experimental question. The simplicity of the neutron-antineutron system made it attractive for the investigation of this phenomenon. In a series of increasingly ambitious measurements at the ILL, the characteristic time (oscillation period) for the neutron-antineutron ($n - \bar{n}$) transition was determined to be greater than or equal to 10^8 s. Such a result is, of course, consistent with baryon conservation.

The $n - \bar{n}$ measurements are also of interest in that they represent the rather singular nature of fundamental cold neutron experiments. The motivation for the $n - \bar{n}$ work arose from the theoretical particle physics community, and initial research proposals developed among high-energy physicists who had little previous contact with the neutron physics community. A rather sophisticated apparatus was constructed *solely for the $n - \bar{n}$ measurement*. This apparatus was installed on a specially prepared neutron beam line. Upon the completion of data collection, this apparatus was dismantled, freeing the neutron beam for other work. The overall program involved many man years of effort. While the scale of the $n - \bar{n}$ program is larger than most other fundamental neutron physics experiments, it nonetheless represents in style, if not in scale, the character of this work.

It is also characteristic of cold beam experiments that their sensitivity is limited by the intensity of the neutron beam available. The actual figure-of-merit (for the neutron flux) for a particular experiment will depend on the nature of the experiment. For example, measurements involving an observation of the decay of neutrons in flight (the neutron lifetime or determination of correlation coefficients) typically require maximum capture flux (flux weighted by the reciprocal of the velocity) within the aperture and solid angle acceptance of the detector. Cold neutrons are clearly desirable due to their low velocity. However, because the divergence of the low-energy neutrons emerging from a guide may exceed the acceptance of the detector, the actual figure-of-merit for the beam will not scale with the "raw" capture flux emerging from the guide. The figure-of-merit for polarized beam experiments will depend on some combination of integrated flux (or capture flux), I , and polarization, P . This may be a simple product PT or in cases where polarization analysis is carried out, P^2T .

While the precise figure-of-merit may vary with the experiment, it is almost universally the case that the *time-integrated intensity* is critical. Thus, the availability of a high-flux reactor with an order of magnitude improvement in beam intensities (such as the ANS) will significantly advance this field. It is less clear that there will be *new* opportunities in this area with a spallation source of the scale under consideration.

4.4.3 Particle Physics with Stored Ultracold Neutrons

Measurement of the neutron electric dipole moment (EDM), d_n , and the beta-decay lifetime of the neutron, τ_n , which are possible using neutrons of extremely low energy (ultracold neutrons⁵⁻⁷ or UCN) are proving to be valuable in achieving a better understanding of the symmetry-breaking components of fundamental forces. The neutron is a composite structure comprising two "down" quarks and one "up" quark bound together by the "strong interaction". In spite of the neutron's electric charge neutrality, the electromagnetic force (10^{-2} times the strength of the strong force) manifests itself with a non-zero neutron magnetic moment. Neutron structure is also influenced by the "weak" interaction which, although its strength is 10^{-7} times that of the strong force, causes the neutron to beta decay. The weak force has "handed" characteristics which lead to the violation of parity symmetry P. (This is not the case for the strong and electromagnetic forces which conserve parity). Neutron decay, like most beta decays, is a parity violating phenomenon. The relative "weakness" of the weak force is responsible for the relatively long neutron lifetime of 888 ± 2 s. In 1964, experiments on the decay of the K^0 mesons revealed another even weaker "force" which violates both parity and time-reversal symmetry T. This force too, which is 10^{-12} times the strength of the strong force, would also be expected to play a role in the internal structure of the neutron. In view of this minute strength, it is perhaps not surprising that other manifestations of this force have so far proved to be elusive. Thus, it has proved difficult to develop a theory for the force, or indeed even to determine whether it is a new interaction or simply a novel manifestation of other interactions, without experimental results in a variety of systems. The simplicity of the neutron makes it an attractive candidate for such a system. The P and T breaking symmetry properties allow the force to generate permanent EDM's in free particles. The size of the EDM, d_n , may be less than or equal to 4×10^{-12} times the diameter of the neutron; that is, $d_n \leq 4 \times 10^{-26}$ e-cm. From experiments with stored UCN, the present 1σ error is 4×10^{-26} e-cm. The strong incentive to improve the precision is immediately evident.

The weak force with a relative strength of 10^{-7} is more accessible, and consequently, it is much better understood. It exists by virtue of exchange of the known heavy W^+ , W^- , and Z^0 particles, and it is related in detail to the electromagnetic force in the theory of Weinberg and Salam. Beta decay is its most direct manifestation at low energies, and the decay of the neutron is the simplest case where quantitative low-energy measurements can be made. The neutron lifetime, τ_n , and the asymmetry parameter for the correlation between the decay electron momentum vector and the neutron spin direction, furnish simultaneous equations which can be solved for the parameters g_V and g_A , which represent the strengths of the vector and the axial vector parts of the weak force for the nucleon. The same value of g_V , with some small corrections, will apply to all nuclear beta decays and to the decay of the muon. The shortage of precision in the neutron measurements has frustrated the pursuit of the finer details of this theory. In the last five years, measurements with stored UCN have improved the precision of τ_n by nearly an order of magnitude, and there has been improved consistency between different experiments. In the future, the asymmetry parameter may also be measured using UCN. Further improvements by a factor of 10 are highly desirable for both quantities, as they will then provide one of the most stringent tests of the standard model for the weak interaction.

Thermal neutron speeds are typically 2000 m/s. UCN have speeds less than about 8 m/s. They are characterized by the fact that they can be stored in "bottles" with material walls. During storage they steadily disappear from the bottle due to beta decay and loss processes involving nuclei in the wall surface. Average storage times of hundreds of seconds can be obtained with clean, room temperature walls of suitable materials. During storage, the neutrons continually bounce to and fro in the storage vessel, reflecting elastically from the walls, and they may cross the bottle 10000 times before being lost. Observation times for individual neutrons in UCN experiments may be hundreds, or even thousands, of seconds. This may be contrasted to observation times which are restricted to tens of milliseconds for cold beam experiments. This huge advantage is partly offset by lower counting rates, but the net improvement in precision can still be two orders of magnitude. In addition, UCN storage vessels can be situated in positions which are displaced by several meters from the source line-of-sight. This results in very low background conditions. Nonetheless, the low absolute count rates associated with all UCN measurements have resulted in the experiments being limited by counting statistics. All UCN measurements will therefore profit from the higher UCN densities which are possible at advanced neutron sources.

At present, the most intense source of UCN is at the ILL. The number density of UCN up to the speed of 6 m/s at the output of the turbine blades is about 90 cm^{-3} . The second most intense source is that at the WWR-M reactor of the St. Petersburg Nuclear Physics Institute (Russia); the density is about five times less, reflecting the comparably lower source thermal neutron flux. These sources have been in use since 1986, and there are no immediate plans (or possibilities) for more intense sources at these institutions. The proposed ANS reactor has a thermal flux at its cold source, which is expected to be ten times higher than at the ILL. This will allow extraction of UCN with a tenfold higher number density. This assumes a similar extraction efficiency of about 8% in each case. The losses result from the product of a factor of 1/6 from reflections in the guide tube and a factor of 1/2 from transmission through safety and vacuum windows.

For an advanced spallation source with ten times the power of ISIS, the potential UCN output is similar to the ANS reactor, but the technology required is not so well tried and tested. It involves a rotor (or reciprocating device) using Bragg reflection from crystals of a synthetic fluorinated mica. The device works with $\sim 10 \text{ \AA}$ wavelength neutrons from the cold source at the peak intensity in the pulse. Since the time-averaged flux at a high-intensity spallation source will be typically about 50 times less than that of an ANS-class reactor, the duty cycle factor *must* be about 1 to 50 to gain complete compensation. This requires a spallation pulse repetition rate of not more than 100 Hz.

Down-scattering in superfluid helium is an alternative method of UCN production which could offer densities of about 10^4 cm^{-3} in the helium, but here, the technology of the device with its requirement for a cooling power of about 1 W at 0.7 K is even more complex. In principle, this type of UCN source could be set up at an existing neutron source and, thus, is not entirely dependent on the introduction of a new source. An advanced neutron source may offer greater flexibility in positioning such a source with a more favorable neutron-to-gamma flux ratio.

4.4.4 Neutron Interferometry

The perfect silicon crystal neutron interferometer has provided the technical means by which increasingly sophisticated quantum mechanical interference phenomena on a macroscopic scale can be observed. For example, the observation of gravitationally induced quantum interference by neutron interferometry remains the only laboratory experiment in which both gravity and quantum mechanics *simultaneously* play an important role. The compendium of fundamental physics experiments carried out with such devices continues to grow.⁸⁻¹¹ It includes the observation of the phase shift of a neutron wave due to the earth's rotation (Sagnac effect), the phase shift due to spin precession, the superposition of spin states (Wigner phenomenon), and more recently, the first observation of the scalar Aharonov-Bohm effect. Devices currently in use are of order 10 cm in length and are sensitive to interaction potentials of the order of 10^{-13} eV.

There is now considerable interest in scaling up the size and, thus, the sensitivity of these devices to search for much smaller and more subtle effects, such as the coupling of the neutron spin to rotation (Mashhoon effect) or performing a neutron (i.e., matter wave) version of the Michelson-Morley experiment. This latter experiment will likely be limited fundamentally by intensity. The reason for this limitation reflects a difference in the geometry of the Michelson-Morley interferometer from those employed to date. Devices currently in use are *achromatic* monolithic devices having an enclosed area (Mach-Zender geometry). Such devices are sensitive to rotations but rather insensitive to translations. However, a perfect-crystal Michelson interferometer is *chromatic*, which results in a counting rate which may be reduced by a factor of 1000 from a comparably "illuminated" classical device. For such an experiment, a neutron source with a flux of 10^{15} n/cm²-s or greater will be essential in carrying out a serious experiment of this type.

Experiments of this kind require an experimental environment which is considerably less "hostile" — in terms of vibration level, temperature stability, and acoustic noise levels — than is common at most current neutron facilities. This is particularly the case if large, split-component devices are to be implemented successfully. In order to facilitate this "quiet" environment, a vibrational isolation structure (i.e., isolated pier) should be designed into the experimental hall at the outset.

Until now, potential applications of the perfect silicon interferometer for materials sciences and solid-state physics have failed to be vigorously exploited. Largely, this is a result of intensity limitations. Two rather attractive ideas which have been suggested for the application of neutron interferometry are *phase contrast microtomography* and *surface "transmitometry."*

The principle behind phase contrast microtomography can be understood by considering the placement of a flat, parallel sample, containing internal density fluctuations (due perhaps to internal interfaces) in one of the beams of the interferometer. These internal density variations will give rise to phase variations across the beam. Upon interference of this beam with the other beam (which maintains a uniform phase profile), these phase variations will manifest themselves as intensity variations across the output beam. Thus, the recombined beam intensity profile will carry

information on the density variations in the sample. This information can be imaged by viewing the recombined beam with a 2-D detector. If a detector with a pixel size of $1\ \mu\text{m} \times 1\ \mu\text{m}$ is used (not yet a technical feasibility), density fluctuations on this scale will be distinctly imaged. Rough estimates give a count rate of one count per pixel per hour with a neutron source having an intensity of $10^{16}\ \text{n/cm}^2\cdot\text{s}$. Clearly, for the successful implementation of this idea to be practical, two developments are required: a very intense neutron source and a position-sensitive detector with very high resolution. The second novel application of neutron interferometry may be understood by considering a flat sample with surface overlayers (of total thickness D) placed in one of the beam paths of a neutron interferometer and oriented near the critical angle θ_c . The phase shift of the transmitted wave,

$$\Delta\phi = \int_0^D \sqrt{q_{\perp}^2 - 2m/\hbar^2 V(z)}\ dz ,$$

is then measured directly. Here, $q_{\perp}$ is the wave vector normal to the sample surface, and $V(z)$ describes the interaction potential of the neutron with the sample as a function of depth. The measurement of this depth-dependent potential $V(z)$ is the goal of all neutron reflectometry experiments. In the interferometric case described above, the phase shift $\Delta\phi$ (which is a function of the angle of incidence) is related to $V(z)$ by a simple integral. This contrasts with the more complicated extraction of $V(z)$ from the analysis of reflectometry data. A measurement of $V(z)$ by this technique (which we call surface "transmitometry") will require a rather narrow (on the order of 0.1 mm), well-collimated (on the order of 1 arc minute) neutron beam. Again, the practicality of this method to materials research will require a very intense source.

An interferometer utilizing very cold neutrons of wavelength $100\ \text{\AA}$ has been successfully tested at the ILL. The neutron optical elements were three high-precision phase gratings with a lattice constant of $2\ \mu\text{m}$, which were used in transmission mode. The prototype interferometer is of length 50 cm, with a possibility of being increased to several meters. This device is very sensitive to external fields, in particular gravity. With certain modifications, it should be possible to detect the presence of nearby massive bodies. Detection of this phase shift may be viewed as a quantum mechanical Cavendish experiment on the neutron.

For all of these neutron interferometry experiments, it is the *time-averaged flux* which is relevant. Thus, a high-flux reactor is preferred over a high-peak-flux spallation source.

4.4.5 Nuclear Physics

Over the last 20 years, there has been a prolific program of (n, γ) nuclear spectroscopy centered on the high-flux reactors at the ILL and BNL. A recent and significant development in this field from the last four years is the gamma-ray-induced Doppler broadening (GRID) method.¹² GRID provides measurements of excited nuclear state lifetimes, a task for which there has previously been a shortage of methods. The GRID method was made possible by the new very high resolution crystal diffraction gamma-ray spectrometer GAMS4, built at NIST and installed in

a collaborative NIST-ILL effort. Neutron capture has been immensely useful in nuclear physics, since it is unselective compared to most other reactions and thus gives access to a wide range of nuclear states. Frequently, capture is followed by a cascade of gamma transitions. The first transition of a cascade is of lesser interest in the GRID method, but it serves to impart a recoil with an energy in the range from a few electron volts to a few kiloelectron volts to the emitting nucleus. The second gamma of the cascade is then emitted from a recoiling nucleus and is subject to Doppler shifts. Depending on the lifetime of the nuclear state involved, the period of emission may embrace both the duration of the recoil and a period when the nucleus has come to rest. The measured Doppler-broadened line shape carries information on the relation between the lifetime and the slowing-down time. The latter, in effect, serves as a clock to determine the lifetime.

A fertile bootstrap process has developed in which the GRID method provides useful information to refine slowing-down theory, and this in turn improves the accuracy of the GRID lifetime measurements. As an example, a cascade in Ti has been studied using both Ti and TiC targets. The change in line shape is then due entirely to the change in the environment for slowing the Ti recoil. There is technical spin-off here since recoil following (n,γ) reactions is an important source of radiation damage in components subject to high neutron fluxes.

As an example of the value of GRID to nuclear structure physics, we cite the recent measurements on the lifetimes in the 1227 and 1234-keV transitions of ^{168}Er . These measurements address the long-standing issue of whether two-phonon collective excitations exist in deformed nuclei. The lifetime result clearly demonstrated the collective nature of these transitions, providing strong evidence for two-phonon states. The GRID technique will continue to be developed and exploited during the next ten years. As is often the case with very high resolution methods, counting rates are a limitation. In this respect, it is not accidental that the work was done at the ILL. The GRID method can certainly increase in scope by accessing weaker transitions should a higher flux neutron source become available.

Some excitement was caused in 1991 by the first resolved measurement of neutron electric polarizability carried out using the pulsed neutron source ORELA. The result claimed is $\alpha_n = (1.2 \pm 0.15 \pm 0.20) \times 10^3 \text{ fm}^3$. The method involves measuring the neutron total cross section of ^{208}Pb from 50 eV to 600 keV. *This work can only be carried out at a pulsed source.* It is important that this measurement be improved further.

Another very active field has been the study of symmetry breaking forces in neutron interactions with nuclei. This dates back to 1964, when Abov and collaborators measured parity violation in γ -ray emission following polarized neutron capture in ^{113}Cd . Interest was raised further by the work of Alfimenkov *et al.* who measured P-breaking asymmetries as large as 0.1 for certain compound resonances. The large enhancement on the base level of 10^{-7} is due to weak-force mixing of nearly degenerate s- and p-wave compound nuclear resonances. With such large effects, it has been possible in a few instances to obtain results for several different resonances in the same nucleus, and then by statistical analysis, to extract the mean-square parity violating matrix element. Neutrons with energies from 1 to 200 eV have been used. This program would benefit from a more powerful pulsed source. At the same time, there have been attempts to measure

parity-breaking effects in more fundamental systems such as the $n(p,d)\gamma$ reaction. Here, the predicted effects really are about 10^{-7} . Related experiments of this kind are the parity-violating spin rotation measurements for neutrons passing through targets of H and He. So far, it has not been possible to obtain resolved results. This program could benefit greatly from the ANS reactor. The study of the recently discovered large parity-violating effects in cold polarized neutron-capture-induced fission would also benefit from the ANS, as would most other fission studies with neutron beams.

The T-symmetry breaking force might be studied by neutron-nucleus reactions exploiting the large enhancements which can occur in compound nuclear states to give a very interesting degree of sensitivity.¹³ The method involves measuring the correlation $\sigma \cdot (I \times p)$, where σ is the neutron spin, p is the neutron momentum, and I is the target nuclear spin. This needs a polarized neutron beam of epithermal neutrons at the energy of the resonance. The highest possible statistics are needed. Such work requires an intense spallation source.

Finally, we would like to add that spallation source installations offer a further range of opportunities for nuclear physics experiments. An unmoderated target can produce beams of neutrons with energies up to several hundred megaelectron volts, offering the possibility of carrying out measurements on neutron-proton bremsstrahlung, neutron-induced pion production, equilibrium and pre-equilibrium emission, fission, and giant resonance properties. These installations also produce neutrinos, pions, and muons.

4.4.6 Conclusions

The development of intense neutron sources has seen the parallel development of a rich research program in which the neutron (and its fundamental interactions) is the object of study. This research, employing a rather wide variety of experimental approaches and techniques, has produced results of importance in particle physics, in nuclear physics, in the determination of fundamental constants, in astrophysics, and in the study of fundamental quantum phenomenology. While the scope and sensitivity of this research has expanded with the refinement of measurement techniques, it is a particular characteristic of this area of study that advances in both the quality and quantity of results have followed directly from improvements in source intensity. It is one of our conclusions that this trend will continue and that this productive area of research will flourish with the next significant advance in neutron source intensity.

The interdisciplinary nature of this research is also manifested in a rather singular characteristic of many of the experiments comprising this work. While most neutron scattering measurements use fixed instruments with rather minor instrumental modification, "fundamental" neutron physics experiments often employ rather complex apparatus specifically constructed for *one measurement*. Such an apparatus sometimes requires many man-years for development and construction (and often very sizable capital investments) and may collect data for a period of weeks. It is quite typical that such an experiment arises from the realization that a tantalizing problem in a rather remote field of physics or astrophysics is best addressed, or even only

addressable, at an intense neutron source. This insight typically develops among researchers outside the neutron physics community. Thus, in a broader view, intense neutron sources act as a valuable resource for a very broad scientific community. An additional conclusion which we draw is that a continued program of important, and often quite unexpected, interdisciplinary research may be expected at an advanced high-flux source.

The diverse nature of the experiments in this area of research is such that it is not possible to state categorically that either a high-flux reactor or an intense spallation source is superior for all classes of measurement. However, there are some general conclusions which may be drawn:

- Research involving cold neutron beams almost always benefits from maximum *time-averaged flux*. The time structure of the beam from a spallation source usually offers no advantage. This work, which would benefit greatly from the availability of a high-flux reactor, will probably not be advanced by the construction of a high-intensity spallation source.
- Both the reactor source and the spallation source will provide the opportunity to increase ultracold neutron densities by a highly desirable factor of ten. The reactor source has the advantage that the UCN production methods are well tried and tested. However, it is possible that future refinements in the production techniques of UCN at spallation sources may yield further increases in UCN density.
- Investigations involving neutron interferometry require the highest possible time-averaged neutron fluxes and thus will be favored by the construction of a high-flux reactor rather than a high-intensity spallation source.
- Nuclear physics research will profit from either a high-flux reactor or an intense spallation source. The character of the research at each source is quite different, and experiments which are feasible at one source may be not be possible at the other. It may be that there is a greater breadth of activities at a spallation source. The two types of sources should be viewed as being complementary. Ideally, both should be available.

4.4.7 References

1. T. von Egidy, ed., *Fundamental Physics with Reactor Neutrons and Neutrinos* (Institute of Physics, 1977).
2. B. Desplanques, F. Gonnewein, and W. Mampe, eds., *Proceedings of the Workshop on Reactor Based Fundamental Physics. J. Physique Colloque C3* (1984).

3. G.L. Greene, ed., *The Investigation of Fundamental Interactions with Cold Neutrons*. Natl. Bur. Stand. (U.S.), Spec. Publ. 711 (U.S. Government Printing Office, 1986).
4. D. Dubbers, W. Mampe, and K. Schreckenbach, eds., *Proceedings of the International Workshop on Fundamental Physics with Cold Neutrons*. Nucl. Instrum. Methods A **284** (1989).
5. L. Koester and A. Steyerl, *Neutron Physics* (Springer-Verlag, 1977).
6. V K. Ignatovich, *The Physics of Ultracold Neutrons* (Clarendon Press, Oxford, 1990).
7. R. Golub, D.J. Richardson, and S. Lamoreaux, *Ultra-Cold Neutrons* (Adam Hilger, 1991).
8. U. Bonse and H. Rauch, eds., *Neutron Interferometry* (Clarendon Press, Oxford, 1979).
9. S.A. Werner and A.G. Klein in *Methods of Experimental Physics*, K. Sköld and D. L. Price, eds., Vol. 23 (Academic Press, 1986).
10. R.J. Birgeneau, D. Moncton, and A. Zeilinger, eds., *Frontiers of Neutron Scattering* (North-Holland, Amsterdam, 1986).
11. G. Badurek, H. Rauch, and A. Zeilinger, eds., *Proceedings of the International Workshop on Matter Wave Interferometry in the Light of Schrödinger's Wave Mechanics* (North-Holland, 1987).
12. R.W. Hoff, ed., *Capture Gamma-ray Spectroscopy*, Pacific Grove, Ca., 1990 (American Institute of Physics, 1990).
13. *Proceedings of the Aqueduct Conference Center Workshop on Tests of Time Reversal Invariance in Neutron Physics* (Chapel Hill, N.C., 1987).

5 User Perspectives

5.1 Perspective of the Neutron Scattering Society of America

H.R. Glyde (University of Delaware)

5.1.1 Summary

This brief summarizes the response from the scientific community to the charge to the BESAC Panel on neutron sources. Views from the community were solicited beginning August 1992. The brief represents a best effort to collate information and views on a short time scale. The eight suggestions for action in Section 5.1.2 address the long-term future source needs, the short-term (seven years) needs (i.e., operation of HFBR, HFIR, IPNS, and LANSCE) and DOE support for investigators.

Neutron scattering is seen as a core competency in materials science and engineering. DOE neutron scattering facilities are a national resource close to the market place. The wide application of materials today rests on the fundamental understanding of structure and excitations in condensed matter revealed by neutron scattering techniques over the past 35 years. The scientific community now uses neutrons both as a unique instrument for innovative discovery and as a key diagnostic tool in research programs ranging from structural biology, polymers, surface science, and superconductivity to metallic defect identification, weld proofing, and clathrate chemistry in oil/water suspensions. Europe has placed a high priority on neutron scattering and, thus, has a spectrum of facilities from the world's best (ILL and ISIS) to small, new and old, with a current total operating budget about twice that in the U.S. and a user community 3-4 times larger.

A Neutron Scattering Society of America (NSSA) survey shows that the scientific and engineering community of neutron scatterers in the U.S. is predominantly young: 85% are under the age of 50. This community has developed in response to the exciting scientific opportunities offered by neutron scattering and to the user programs at DOE and Department of Commerce (DOC) facilities which have made experimentation possible. The DOE support of individual investigators and of the neutron scattering facilities can take credit for the enthusiastic community of neutron scatterers that now exists in the universities, corporations, and national laboratories.

The size and vitality of this community is now limited by support of investigators and the availability of neutrons and instruments. The past three years have seen extended shutdown of some facilities and uncertain operation of others. Without new instrumentation for existing facilities and immediate construction of new facilities, the future prospects for DOE facilities and, therefore for the neutron scattering community, are poor. Submissions to the NSSA voice grave concern. We are at a critical turning point for the future. A key competency could be seriously downgraded or lost. The scientific community shares the energy and enthusiasm at DOE facilities for new instrumentation and new facilities to hold our quality edge.

The scientific community sees reactor-based and spallation-based neutron scattering laboratories as complementary and mutually supporting. Each has unique capabilities given current instruments. This is the central and uniform message from the scientific community. The user community wants access to both type of facilities. How do we get there?

To meet short-term demand, over the next seven years, the community suggests continued operation of all four DOE facilities: HFBR and HFIR, the reactor-based facilities; and IPNS and LANSCE (if possible), the spallation-based facilities. They are all over-subscribed, some by a factor of 3. There is an acute need for new instruments at all the facilities. There has not been the investment in new instruments at DOE facilities during the 1970s and 1980s that has paid rich dividends in Europe (at ILL, ORPHEE, and ISIS). These new instruments can also serve as a stepping stone into new facilities. Extension of the running period of IPNS was proposed. Many recommend a minor share in ISIS and/or the ILL, the world-leading facilities, for U.S. scientists and engineers to meet the predicted shortfall. At present, U.S. participation is 9% at the ILL and 4% at ISIS. The next seven years will be critical to the scientific community.

If the DOE cannot sustain the operating costs of this program, the community proposes the DOE examine the reasons for the sixfold increase in operating costs at HFBR and HFIR since 1977 and make comparisons with NIST. If this does not produce savings, closure of one each of the reactor and spallation sources would at least meet the desire for access to each type of facility. This would make access to ISIS and the ILL even more important. Recall that current operating costs of the ILL and ISIS alone (\$90M/yr) is greater than that of the current U.S. program — HFBR, HFIR, IPNS, LANSCE, MURR, and NIST — (\$80M/yr).

For the long term, if one U.S. facility must be selected, the community proposes proceeding directly with the ANS, combined with clear support for a broad range of instruments. This will take advantage of development of existing expertise in reactors since 1984 and offers the best chance of one of the needed facilities within seven years. The community suggests that the DOE open discussions immediately with Europe on a truly international spallation-source laboratory. Collaboration to explore together imaginative new instrumentation can begin now. Specific development of the project can begin now at a DOE laboratory. The spallation source instrumentation can be used at the international facility — or at one in the U.S. should the two facility option eventually be acceptable. Present facilities would close when the new facilities come on line.

In short, the community sees reactor and spallation sources as complementary and wants access to both. In the long term, this can be best achieved by constructing the ANS now and pursuing a spallation source through international collaboration.

5.1.2 Suggestions for Action

5.1.2.1 The Long Term

The DOE should construct the ANS as now developed rapidly with a modern array of instrumentation.

The ANS was initially proposed in the Seitz-Eastman report of 1984.¹ The need for a new reactor-based neutron scattering facility has only increased since that time. The ANS has now seen eight years of development. Approximately \$60M has been devoted to its design and testing of the design. The ANS is now a clearly articulated project. The community supports the ANS taking advantage of development time and expertise gained over the past eight years.

The ILL has a state-of-the-art reactor of the 1960s. The ILL is successful now and will be in the future because this reactor was followed by a rich, varied, and continued development of instrumentation through the 1970s and 1980s. The ILL will be the leading reactor-based neutron scattering laboratory in the world during the late 1990s and 2000s chiefly because of the instrumentation. A current generation reactor (of the 1990s), with similar adequate funding for new instrumentation, can be a world-class reactor-based neutron facility well into the next century. If a significant saving can be achieved, by reducing the power and/or flux of the ANS (by 2 or 3), the community still supports the ANS, since with imaginative instrumentation, much of this loss can be recovered. The community is not able to evaluate whether a significant saving is possible.

Essentially, the neutron scattering community is seriously concerned about any further delay of new neutron facilities. Given the complementary nature of reactor and spallation sources, the NSSA suggests construction of a reactor-based neutron scattering laboratory, now taking advantage of existing development and current expertise in reactor technology.

The DOE should begin discussions for a new spallation neutron source based research laboratory, preferably in collaboration with Europe and/or Japan.

Given the complementary nature of spallation and reactor sources, there is also a need for access to an advanced spallation neutron source. The projected cost of the ANS is high. The Advanced Photon Source is expected to cost \$792M. These costs signal a new era, an era in which the U.S., on its own, clearly cannot afford to construct one of all types of facility. Discussion of a new European spallation source (ESS) has already begun. The community suggests that the DOE begin planning for a new spallation source in the 1-5 MW beam power range in collaboration with Europe. This will also take advantage of expertise in Europe with ISIS. Design and development of a source can begin at a DOE laboratory, now taking advantage of U.S. experience with IPNS and LANSCE. Collaboration on new instrumentation can begin immediately with ISIS. From lessons learned by the High Energy Physics community, major

facilities are more likely to be successful when conceived as collaborative projects from the beginning.

These recommendations should really be taken together. It might be reasonable to elect to construct a U.S. neutron scattering laboratory based on a spallation neutron source now. In this case, participation in a new "international," world-class reactor-based facility should also be pursued. Given the U.S. lead with the ANS and current discussions of the ESS in Europe, the community believes the order proposed here offers distinctly the best route to a new world-class U.S. neutron facility plus collaboration in and access to the other. Time and an international context are of the essence from a user perspective.

5.1.2.2 In the Shorter Term

New instruments at existing DOE facilities.

European facilities have had an extensive program of spectrometer development and construction during the 1970s and 1980s. ILL has approximately 50 instruments in total; ORPHEE, 25; and ISIS, 13. This is nearly twice the combined number at HFBR, HFIR, IPNS, and LANSCE in the U.S. Many instruments at DOE reactors are 25 years old. Instrumentation is needed at spallation sources both now and for future sources.

The community supports a major program of instrument building at all DOE facilities. This would both upgrade the quality of facilities in the short term and establish a competency and prepare the way for instrument building for the new sources. The community believes there has been too little emphasis placed on instrumentation and associated operating costs.

Extension of the operating cycles at LANSCE and IPNS to full-year operation if possible.

This will:

- increase the availability of neutrons in the 3–7 year time scale for the scientific community.
- provide experience (at IPNS and LANSCE) with intense spallation sources in preparation for a new source.

In 1990 and 1991, LANSCE operated only a few weeks per year. In 1991, IPNS operated 16 weeks. Approximately one proposed experiment in three can be allocated beam time at IPNS. The planned operation of LANSCE for only four months per year leads to a large ratio of start-up-problems-to-smooth-operation and is therefore very inefficient. For example, at ISIS, the beam is down 50% of the time in the first cycle after annual shutdown.

The community would like to see adequate funding to increase the weeks of operation per year. This is not simple at LANSCE. However, many submissions from the community expressed clear and articulate support for LANSCE. Their research programs depend on it. The community foresees a major shortage of spallation source neutrons in the short term.

The DOE should consider a minor share in the ILL and/or ISIS.

The purpose is to increase the availability of neutrons and access to many new instruments over the next few years. The community sees a serious shortfall.

At present, ISIS and the ILL are open to use by U.S. citizens and residents on a proposal basis. This is particularly true if the experiment is done in collaboration with a European. However, for many users, getting started is a major problem. Financing international travel and a stay of several days in Europe is also a serious problem. There are many barriers. Current use of ISIS by Americans is 4%, and 9% at the ILL.

A share in the ILL, for example, would open the door to exclusively U.S. proposals. It would also provide travel and accommodation funds to do the experiment once the proposal is accepted. Instrument responsables and graduate students can be supported at the ILL.

Rather than a share, a program to assist use of the ILL and ISIS through specific funding for travel and other costs could be considered.

The DOE should consider upgrades to small research reactors in universities to explore whether these could become part of a network of neutron scattering centers.

The European network of neutron scattering facilities consists not only of the major facilities, such as the ILL, but also of many lower intensity research reactors. Experiments can be tested for feasibility at the less intense neutron sources where there is less pressure on time. When tested, the experiment can be completed at a major facility with final data taken there. The NSSA appreciates that it may not be rewarding to upgrade many of the smaller research reactors.

5.1.2.3 Support for Investigators

The DOE should provide funds to DOE user facilities for support of user travel and personnel costs at the facility such as at the ILL.

At any meeting of the community, the difficulty of supporting experiments and personnel at the facility and travel to and from the facility is raised. This seems particularly true for the new user or for new experiments, novel ideas that are risky and are not yet supported. This issue is not fully understood, but is a cry from the community. There are clear models for support of user

travel and other expenses at neutron facilities in Europe. The NSSA suggests provision of funds for user support to be allocated by the DOE facility on a discretionary basis when proposals are accepted.

The DOE should ensure support of individual research programs.

The foundation of all these issues is the support of individual research programs by the NSF, the DOE, and other agencies. The vitality of the user community rests on the DOE support of investigator research programs through its Office of Basic Energy Sciences (BES). This point is emphasized in the Seitz-Eastman report.

5.1.3 Neutron Scattering Society of America

The following sections are taken from a brief prepared by the NSSA, a fledgling society formed in January 1992 to foster the use of neutron scattering in science and engineering and to represent the community of scientists who use neutrons. The NSSA now has 350 members.

In late July 1992, the BESAC Panel asked the NSSA to seek views of its members on neutron sources. Over 50 written statements, some running to several pages, were received. Approximately 100 telephone discussions were held. Sections of the brief were circulated for comment to those who sent written statements. The brief was prepared on too short a time scale to develop a consensus. The process was imperfect, but we believe the suggestions for action reflect, broadly, the views and priorities of the community. The brief does not address specific issues of cost, technical and environmental feasibility, political, and organizational factors which enter the decision of choices of neutron sources and instrumentation.

5.1.4 Nature of the Neutron Scattering Community

The community of scientists and engineers who use neutrons in their research is predominantly a young community. The NSSA undertook a study of the age distributions of neutron users at three of the major facilities during 1991. The facilities were CNRf at NIST, the IPNS at ANL, and LANSCE at LANL. These facilities were selected chiefly for regional distribution throughout the U.S. The CNRf is a reactor-based neutron facility, while IPNS and LANSCE are spallation-source-based neutron facilities. BNL has also recently participated in the survey. The attached age distribution chart (Figure 5.1) shows clearly that the community is predominantly young. Some 85% of users are below the age of 50, and 60% are below the age of 40. The users are from universities, corporations, and the national laboratories. Approximately 650 users are included in the survey. Some users did several experiments but were counted only once. Graduate students and postdoctoral fellows are included in the survey.

FIGURE 5.1 Age Distribution of the U.S. Neutron Community.

The neutron community falls approximately into two parts. The first part is small and made up of the older, professional neutron scatterers. Many of these individuals are located in the national laboratories, some in universities. The scientific contributions of these individuals, who have made a career in neutron scattering, are readily identifiable. Their achievements have laid the foundations of condensed matter physics and materials science. Their work has stood the test of time. The second part is a much larger, younger group of scientists and engineers who have come into neutron scattering more recently as a result of the user programs. While the former group is predominantly in condensed matter physics, the second group is spread over many fields, from biology to engineering. It is more difficult at this time to identify their scientific achievements because these achievements are both spread over many disciplines and have not had the test of time.

The size of the young portion of the neutron scattering community is strictly limited now by the availability of neutrons. For example, at IPNS approximately one proposal in three for an experiment is accepted. Comments from individuals received for this brief show there is great difficulty in obtaining neutrons for experiments, generally throughout the community. As an example, one individual had to try several sources over a period of two years to do a structural study of polymers. The community is therefore seriously concerned about the present availability of neutrons.

A second concern are funds to travel and execute research at the neutron facilities. This is not so much a problem for the older scientists, but it is certainly a problem for younger individuals coming into the field. This serious concern over the availability of neutrons and funding to perform experiments dictates our recommendations. We note at this point that the availability of neutrons has not increased over the past 15 years (see Table 5.1).

A critical issue is the perception of neutron availability for the coming three to seven years, given the age of DOE facilities and much of the instrumentation. This is a most critical issue since the young component of the community must demonstrate scientific achievement on this time scale. This demonstration is necessary both to secure funding for research and to secure tenure and promotion as university professors. While the choice between reactor- or spallation-based sources and construction is clearly a longer time scale than this, the three to seven year time scale is important for the survival of the community. If the younger participants feel they cannot find achievement in a three to five year time scale using neutrons, they will simply turn to other research.

5.1.5 Achievements and Opportunities Using Neutrons

The driving scientific interest in neutron scattering in the community is based on the rich history of scientific discovery in the past, perceived opportunities in the future, the use of neutrons as a diagnostic tool in a broad spectrum of fields, and the growing applications in product development and testing. Europe has clearly assigned a high priority to neutron scattering. These points are sketched below.

TABLE 5.1 Costs and Availability of Neutron Scattering Sources

<i>Reactors</i>								
Facility	Operating ^a Cost		Availability ^b					
			Thermal Flux (10 ¹⁴ n/cm ² .sec)		Number of Spectrometers		Operating Days per Year	
	1977	1991	1977	1991	1977	1991	1977	1991
HFIR	4.1 \$M	27.3 \$M	13	11	8	9	~330	210
HFBR	2.8	20.7	8	5	12	13	~270	60 ^c
NBSR	1.2	5.5	2	4	11	12	~300	296
MURR	0.8	4.0	1	1	8	8	~330	~330
HFR (ILL)	90 M FF	315 M FF	12	12	29	32	~260	~260 ^d
<i>Pulsed Sources</i>								
Facility	Operating ^a Cost		Availability ^b				Reliability	
			Avg. Proton Beam Power (kW)	Number of Spectrometers		Scheduled Days per Year		
	1991		1991	1991		1991	1991	
LANSCE	14.3 \$M		38	8		37 ^e	59%	
IPNS	6.7		6	12		112	95%	
ISIS	~15 £M		77	13		167	78%	

^a Not all are reported on the same basis. In some cases, part or all of the user program and/or the in-house scientific programs are covered in the specified operating cost.

^b This should not be viewed as a complete basis for intercomparison of facilities, as other factors such as the availability of cold moderators, locations and areas of moderator viewed by beam tubes, type of target (in the pulsed-source case), etc., also figure into the overall effectiveness of the facility.

^c Only operated 4.5 months in 1991. Normal operation ~265 days per year.

^d Nominal full-year operation schedule before reactor shutdown in May 1991.

^e Scheduled to operate 4 months per year at same cost.

In 1954, Placzek and van Hove first proposed that phonons in solids could be observed by neutron scattering. Our current understanding of materials, from simple solids to complex biological structures, now rests on the fundamental discovery and display of structure and excitations in matter using neutrons. This historical discovery ranges from Landau rotons, magnons, and magnetic critical phenomena to van Hove singularities and Kohn anomalies. This rich display would not be possible, even today, without neutrons.

Over the last ten years, any list of the most significant achievements would reflect the following characteristics. First, the breadth of neutron scattering use has increased dramatically. SANS has been steadily applied to metallurgical, polymeric, and biological systems. Applications in structural biology have grown. Reflectometry has been successful. Perhaps the breadth is best reflected by the new area of neutron residual stress measurements in engineering materials and components, which has sufficiently developed to be a source of revenue at some sites. Thus, physical metallurgists and welding engineers, as well as physicists, are visiting reactors to discuss neutron stress measurements. Second, neutron scattering has spurred key developments in physics (e.g., high-temperature superconductors, where primary magnetic and structural information has been supplied). Third, the more traditional areas of neutron scattering have continued to develop and deepen. Neutron interferometry, magnetic scattering, and lattice dynamics have made significant advances. Fourth, new instruments have continued to be developed, as well as monochromators and detectors.

In reviewing the record, perhaps the most notable achievement of recent years has been the creation of new opportunities. The question now is how best to deliver the potential of the method to the growing community of interested users. The record also shows that the growth of the field and the development of the neutron user community in the U.S. has occurred through use of facilities that may be world-class but are clearly secondary to sources now in place in Europe. New generation reactor and spallation sources are going to be required to keep the initiative and exploit the potential that has energized the growing user base. More important in the interim period, improvement in currently successful facilities, including university research reactors, is the most cost-effective way to realize the opportunities that have been created.

What are the future opportunities? It is helpful to note that materials and other research done using neutrons is not like particle physics. In particle physics, the key experiments are highly focused and well known: to observe the top quark, the Higgs Boson, departures from the Standard Model. These are known years in advance. In contrast, the contributions of neutron scattering are spread over many fields: biology, pharmacology, polymers, chemistry, materials and product testing, etc. Fields change rapidly and emerge suddenly. Who would have predicted the central role of neutrons in superconductivity before 1986 or the current wide use of neutrons in polymers?

It is, therefore, very difficult to predict opportunities in neutron scattering.

Rather a case can be based on the energy and intellect of a growing community of scientists and engineers who seek neutrons and a fine historical record.

As noted, the scientific community using neutrons is predominantly young. Many of the younger members use neutrons as a diagnostic tool, among others, in their research. As well as a unique tool for discovery, neutron scattering is now used as an instrument to characterize materials in the widest sense of the word materials (e.g., in structural biology). Much of this is close to the market place. Neutron scattering is therefore a national resource in materials research and product development for the market place. Materials is a national priority. This is an important new role for neutrons, well recognized in Europe.

How much funding should be devoted to neutron scattering? The combined countries France, Germany, and the United Kingdom form a block of comparable population and Gross Domestic Product (GDP). Using Tables 5.2 and 5.3, we see that current operating expenditures devoted to the ILL and ISIS alone exceed that currently at DOE neutron facilities. Including the

TABLE 5.2 Summary of User Statistics for FY 1991

	IPNS	LANSCE	HFBR ^a	HFIR
No. of Users Total	236	79	168	95
University	110	34	99	41
Laboratory/Government	24	15	28	18
Industry	75	20	23	4
Foreign	37	10	18	22
No. Beam Lines	12 instruments	10	9	6
Average Time at Facility	4 days	9 days	7 days	7 days
Operating Expense (\$M)	6.725	14.264	20.722	27.350

^a Restart May 1991.

TABLE 5.3 European Major Facilities

	ILL	ISIS
Flux	$\sim 10^{15}$ n/cm ² .s	60 μ A
No. of Instruments	35	14
Annual No. of Users (approximate)	2000	750
Annual Operating Expense (\$M)	60	30

other European facilities in Table 5.4, the current operating expenditures in Europe are roughly double that in the U.S. It would be interesting to make this precise. On these grounds there is no case for reducing the operating funds to neutron facilities in the U.S.

Europe has particularly devoted substantial funding to new neutron spectrometers and direct support of the scientific community using neutrons through their respective scientific councils. As a result, the neutron scattering community in universities, corporations, and national laboratories in Europe is at least three times larger than that in the U.S. The short deadline for this brief precluded making this more precise. Particularly, the role of neutrons as an incisive tool to characterize materials is well recognized and vigorously supported in Europe.

Europe has clearly identified neutron scattering as a high priority. On a basis of comparison, operating support for facilities and support for investigators should increase substantially in the U.S.

5.1.6 Complementarity of Reactor and Spallation Source Based Facilities

The present charge to the BESAC Panel on the future of neutron scattering in the U.S. is focused on the relative merits of reactor-based neutron sources and accelerator-based spallation neutron sources. From the view of the users, the two sources are complementary. In terms of the scientific opportunities, the sources are complementary. While there are fields of study and experiments in which either can be used, there are specific domains in which one is significantly better than the other. It is for this reason that Europe has a major facility of each type: the ILL and ISIS. It is for this reason that Europe is considering both types of sources in its future plans, with a spallation source taking the lead. If the U.S. must opt for one source type, the user community would like to have access to the other on an international collaboration basis. Users made the following points:

5.1.6.1 Beam Characteristics

The thermal neutrons from a steady-state reactor are largely confined to low momentum and energy. The beam can be characterized as confined in Q and ω and is continuous in time. If a cold source is added, the beam can be concentrated to very low Q and ω .

In contrast the beam from a spallation source is spread over a wide Q and ω range. Particularly, it has a long tail extending up to high energy. The beam is pulsed and good for TOF instruments.

TABLE 5.4 European Neutron Scattering Facilities

UNITED KINGDOM	
(a)	ISIS – Current $\approx 150 \mu\text{A}$; 14 instruments; Users ≈ 750 per year; Operates ≈ 200 days.
(b)	Contribution to ILL.
GERMANY	
(a)	Contribution to ILL.
(b)	JULICH Reactor – Flux $\approx 1.5 \times 10^{14} \text{ n/cm}^2\cdot\text{s}$; 14 instruments; Upgraded with cold source and instrument hall; Comparable to CNRF, NIST.
(c)	HMI, Berlin – Flux $\approx 10^{14} \text{ n/cm}^2\cdot\text{s}$; 12 instruments; Upgraded with cold source; Currently running at low power; 70% of NIST, potential to be comparable.
(d)	Munich and Geestbach Reactors – Both small, done pioneering work (e.g., first SANS experiments); Few instruments; No user program, but available for test experiments.
FRANCE	
(a)	Contribution to ILL.
(b)	ORPHEE, Saclay – Flux $\approx 3 \times 10^{14} \text{ n/cm}^2\cdot\text{s}$; 26 instruments; Cold source and extensive guide hall (impressive instrumentation); Operates 250 days per year; 120% of NIST.
(c)	SILOE, CENG, Grenoble – Old reactor; Good flux; 8 instruments; Operates 200 days per year; A few users; Done some excellent experiments; May close in 2–3 years.

5.1.6.2 Fields of Overlap

With proper instrumentation adapted to beam characteristics, both sources can be used for many experiments. One example is diffraction and structure studies of amorphous materials, long-chain molecules, large biological structures, and polymers. Explicitly, SANS can be done readily with both sources.

5.1.6.3 Reactor Source Advantages

A reactor source is far superior for experiments needing *low-energy and high-Q resolution*. This is achieved with reasonable flux using a beam of limited Q and ω range. In many studies, high- Q resolution is critical. Examples, are low-lying excitations in crystals and other samples in which the dispersion curve ($\omega(Q)$) is rapidly varying with Q . In these cases, high definition in reciprocal space is needed. This is especially achievable if the reactor has a cold source. In principle, the dispersion relation can be measured with a pulsed source, but the practical difficulty

is that the experiments are generally carried out at constant angle, not constant Q . Numerical unfolding has to be performed to carry out the analysis. Thus, the raw data is more difficult to assess in order to alter the experimental protocol in real time. The time-averaged flux is what is important, which is most economically provided by a continuous reactor beam.

Today, the only polarized neutron sources are available at reactor sources. NSE spectrometers currently exist only at reactor sources.

5.1.6.4 Spallation Source Advantages

Large Energy Transfer. When inelastic events are to be studied with energy transfers greater than about 100 MeV, the spallation source is the only practicable choice because of the high-energy tail in the distribution. These situations arise, for example, in molecular vibrational spectroscopy, in high-energy magnetic excitations, and in deep inelastic scattering measurements of momentum distributions. In some cases in this energy range, there are other probes (e.g., infrared) which are cheaper and easier to use. However, because of the weak nature of the neutron-matter interaction and because neutrons interact with almost all nuclei with advantages in selection rules and isotopic substitution, neutrons have broad application and are unique in many cases.

Isotropic Systems. For q independent situations (e.g., molecular energy levels) or scalar q problems (e.g., amorphous or liquid state dispersion laws), the higher data collection rate (all q 's simultaneously) associated with spallation sources is an advantage provided one has large area detectors. The same gain exists for neutron reflectometry.

5.1.6.5 Other Findings

The committee report to the Science Board of the SERC in the U.K., chaired by P.H. Fowler (1990), examined the scientific opportunities offered by the ILL (reactor-based) and the ISIS (spallation-based) neutron scattering laboratories. They concluded that the ILL and ISIS were complementary and both should be supported.

The response from the community for this brief is approximately equally split between reactor and spallation sources. Many recognize the advantages of each and many use both types of facility. The advantages can be, and certainly will be, exploited by new advances in instrumentation.

The suggestions for action proposed here are based on the complementary nature of the two sources as seen by the community and from the perspective of scientific opportunities.

If both sources cannot be built in the U.S., the community would like to see one built now with the other available through an international collaboration. Critically important is a decision to proceed now, without delay, on a new neutron scattering laboratory including instrumentation.

5.1.7 Neutron Availability in the U.S.: Comparisons with Europe

5.1.7.1 Neutron Availability in the U.S.

In 1977, the National Academy of Sciences (NAS) issued a report on the state of neutron scattering in the U.S. (*Neutron Research on Condensed Matter: A Study of the Facilities and Scientific Opportunities in the United States*). Included in that report was a table of U.S. neutron scattering facilities and some of their relevant parameters. At that time, there were four major reactor facilities used for neutron scattering in the U.S., as well as a number of smaller, less-well-instrumented reactors. These four major reactor facilities are still operational, and Table 5.1 compares some of their parameters in 1977 with the corresponding values for 1991 (the last full year for which data are available). It can be seen from the table, that with one exception, the only parameter in which there has been a significant change over the 14-year period is the cost of operation of these facilities. The major exception is the NBSR reactor at NIST, which has undergone a doubling of operating power during this period and has also added a guide hall which is just now being instrumented. During this same period, 4 of the 9 smaller reactors listed in the NAS report have been shut down. *The net result is that there has been little, if any, increase in the availability of reactor neutrons for neutron scattering science during this period, despite the sixfold increase in the cost of operating the DOE reactors for neutron scattering.* (Inflation over this period has been roughly a factor of 2.5, with the additional operating costs being largely attributable to changes in safety and environmental regulations.) The DOE might review safety, asking whether a given regulation is relevant to a specific facility, and compare with NIST.

However, this same period has seen the introduction of the pulsed neutron sources IPNS at ANL and LANSCE at LANL. Similar parameters on operating cost and availability are shown in Table 5.1. Because of the vast differences in the types of capabilities offered, it is difficult to compare directly the availability of pulsed-source and reactor neutrons for neutron scattering science, but it is clear that the pulsed sources have provided some increase in this availability. It is also clear that this increase could be significantly larger if the pulsed sources were to operate a greater fraction of the year.

The recognized world-leading facilities for both reactor and pulsed-source neutron scattering are located in Europe: the HFR reactor at the ILL in Grenoble, France, and the ISIS pulsed-neutron scattering facility at the RAL in the U.K. Parameters for these facilities are shown in Tables 5.1 and 5.3 for comparison. It can be seen that they significantly exceed the capabilities of any of the U.S. facilities in both the reactor and pulsed-source categories. A number of medium-flux European reactor facilities are also quite active and well-instrumented for neutron

scattering (see Table 5.4), so that the total availability of neutrons for neutron scattering science in Europe is considerably greater than that in the U.S.

5.1.7.2 Comparison with Europe

The three European nations, the U.K., France, and Germany form an advanced nation block with population and GDP comparable to (somewhat less than) the U.S. How does the U.S. compare, in the use of neutrons by the scientific community, with this European block? Does this comparison provide a measure of how much funding might be devoted to neutron scattering? Below we present some very approximate comparisons.

In Table 5.2 are listed the number of users, instruments, and operating expenses of the four major DOE neutron scattering facilities: IPNS, LANSCE, HFBR, and HFIR. (The NSSA is grateful to Mr. F.A. Koomanoff, DOE, Washington for providing the information on which this table is based). The first two (IPNS and LANSCE) are spallation-source facilities, and the second two (HFBR and HFIR), reactor-source facilities. In Table 5.3 are comparable data for the two major and largest facilities in Europe: ILL (France, Germany, and the U.K.) and ISIS (the U.K.). The combined users program at the ILL and ISIS was 2800, compared with 600 for the four DOE facilities. The respective annual operating expense was \$90M, compared to \$70M.

The first point is that the access and user program is approximately four times larger in Europe. In 1991, NIST accommodated approximately 600 users. This reduces the ratio to 2.5. The second point is, that for a comparable operating cost, the two major European facilities accommodated four times as many users, which at the ILL included full travel and accommodation costs for all users from the U.K., France, and Germany. This difference, in our view, is not the fault of the U.S. facilities and is discussed below.

In terms of quality of facility, the ILL, HFBR, and HFIR have comparable neutron beam flux. However, ILL had a continuing program of instrumentation development and building through the 1970s and 1980s, which gives it now an unmatched array of instruments. There is a total of some 50 instruments at the ILL, with 35 in the user program. World-class science and engineering can be and is done at HFBR and HFIR, but not on the scale as done at the ILL.

IPNS pioneered spallation sources in the world and the science done with them. It had the first formal user program in the U.S. The current at IPNS is $\sim 15 \mu\text{A}$ which is now 1/10 that of ISIS. At LANSCE, the usable current is debated but is between 1/3 to equal that of ISIS. In 1991, IPNS operated for 16 weeks, and LANSCE operated for six weeks (the scheduled LANSCE operation is four months per year, and 1991 was a bad year). ISIS operates for approximately 35 weeks a year. The difference in operating time largely accounts for the difference in number of users per year at each source. An overall measure of quality and size of facility is the total beam current (in mA) times operating time (mA-hr) times the number of instruments. In 1991, this figure-of-merit was: ISIS, 5000 mA-hr times Instruments; IPNS, 800; and LANSCE, 600.

Europeans user programs began well before those in the U.S. This developed a community of scientists in the universities interested in neutron scattering. This and the relatively small number of instruments at HFBR and HFIR account for the large difference in number of users between the ILL and HFBR/HFIR.

Finally, the user community expressed some concern about cost of operation, particularly of HFBR and HFIR. The operating costs, compared to CNRF at NIST (\$5.5M in 1991) and the ILL, relative to the number of users is very high. This may be due to the age of the reactors and to new safety regulations implemented by the DOE. In comparison with NIST, the cost of these safety procedures is apparently very high. The DOE might ask whether any individual regulation is relevant to a specific facility. The NSSA is concerned with cost because the ultimate result may be loss of neutrons.

ISIS and the ILL are relatively new facilities. The prospects for the next ten years at these facilities (after 1994) are excellent. The same cannot be said for the DOE facilities — in terms of age (HFBR, HFIR) or performance (IPNS). LANSCE is clearly uncertain at this time. The neutron scattering community is gravely concerned over the availability of neutrons for the coming three to seven years. This concern is turning many away from neutron scattering, especially the young. There is a critical need for new instruments at existing facilities or facilitating use of the ILL or ISIS to provide facilities on three to seven year time frames, as well as starting on an advanced new neutron scattering laboratory immediately to meet the long term need. Other European facilities are listed in Table 5.4.

5.1.8 Reference

1. National Research Council, Major Materials Facilities Committee, *Major Facilities for Materials Research and Related Disciplines*, F. Seitz and D.E. Eastman, Co-Chairs (National Academy Press, Washington, D.C. 1984).

5.2 University Education, Research, and Reactor Community Perspective

J.J. Rhyne (MURR)

The comments and opinions given in this report represent the viewpoints solicited from a group of university scientists, from whom 14 written replies (plus 1 by telephone) were received. In addition, about the same number of informal comments were communicated by fellow scientists at the University of Missouri Research Reactor Center. Although the responses appear to be representative of the larger university community, no attempt was made to make the list comprehensive. The persons responding represented scientists associated with one or more of the following three areas: (1) education of neutron scientists and engineers, (2) performance of neutron scattering and related research in a university environment, and (3) work experience at university (or non-user facility) research reactors.

The university scientists and engineers responding are involved in one of four principal disciplines and associated techniques including (1) neutron scattering (the predominant number), (2) biology and radiochemistry utilizing isotopes produced by university reactors, (3) trace element studies by activation analysis of (often) short-lived isotopes, and (4) nuclear engineering including depth profiling and prompt-gamma ray studies. A summary of the research work performed by the university community is contained in a 1988 NAS report.¹

The university community is characterized by its diversity of research uses of neutrons, ranging from neutron scattering to archaeometry and radiopharmaceutical research, and including materials testing, nutrition and epidemiology studies, fundamental physics, and nuclear engineering. This broad and diverse spectrum of interests places rather stringent requirements on the types and versatility of neutron sources needed to satisfy the university community.

This group is unanimously convinced of the importance and vitality of neutron research and of the major scientific impact that neutron scattering and related work is making in a continually increasing variety of disciplines. This is illustrated by the chart of papers in the last five international conferences on neutron scattering (Fig. 5.2).

A major concern is the loss of U.S. influence in this field to Europe and Japan, particularly since neutron scattering originated here. This loss is a result of the protracted U.S. budget drought for new equipment and facilities. Figure 5.3 illustrates this geographic polarization over time. *A new national neutron source and a renewed national commitment to other aspects of the field, particularly in the education of new scientists, are essential for the future viability of the field.*

FIGURE 5.2 Subject Matter of Papers in Past Five International Conferences on Neutron Scattering.

FIGURE 5.3 Origin of Neutron Scattering Papers Presented during Past Five International Conferences on Neutron Scattering.

The university group represents the prime source of these future scientists, and its perspective is strongly tilted toward facilities that offer the best platform for graduate education. The group is particularly strong on the "feeder reactor" concept that is so highly successful in Europe. This encompasses a group of medium-power regional sources having excellent instrumentation with ready access to the major central facilities (the ILL and ISIS).

The consensus is that the most effective and efficient arrangement is to have a group of well-instrumented local neutron sources on which to train students and perform research not requiring the highest fluxes. As is amply demonstrated at present, these regional sources alone are fully capable of performing highly competent and significant, publishable research, but not of performing those problems requiring the most specialized facilities. Research started at the "feeder" sources can then be transferred to the "finishing school" national centers having higher flux or more specific instrumentation. The environment of the regional reactors (vis à vis the national user facilities) often provides a superior environment for M.S. and Ph.D. research because of less rigid instrument scheduling, enhanced instrument flexibility, and available local supporting research capabilities. Feeder reactors also provide a currently available, near ideal proving ground for new instrumentation concepts to be later transferred to the national laboratory. Costas Stassis (Iowa State University) summarized this feeling: *"In my opinion, the best training for students is done in small University reactors. Their education in scattering techniques can be completed best in a steady-state national neutron facility."*

The idea of having only the national source available requiring extensive and expensive travel and time away from the home institution is highly unpalatable to many university researchers. This is added to the delays inherent in a proposal system and the resulting rigid time schedule for experiments. If only a remote national source were available, the net effect would probably be to diminish the number of university faculty whose *major research focus* is on neutron science, and replace this focus by research using neutrons only peripherally or as an adjunct to other research emphases. The analogy in the x-ray community would be the NSLS or the APS in the absence of local rotating-anode machines.

Reliability is a major concern of having "all the neutron eggs in one basket" at a single national source. Shutdowns, such as have occurred recently at HFBR and HFIR in the U.S. and the ILL in Europe, can only be tolerated if alternative sources for carrying on the research are available.

5.2.1 Recommendations and Requests

5.2.1.1 The National Source

Because of the very diverse scientific interests and expertise of university researchers, the balance strongly favors a new national *reactor* source if only one new national source is to be built. At a slightly lower level, support exists for also building a new generation pulsed source for

scattering research. Otto Harling (Massachusetts Institute of Technology) reflects the general opinion: "... *the training and experience of young scientists who work at university reactors is more transferable over a wide range of research applications to a steady-state ANS rather than to a pulsed ANS,*" which is echoed by Myron Salamon (University of Illinois-Urbana-Champaign): "*In my opinion, the nation needs both a powerful and reliable pulsed source, and one or more high-flux reactor based systems.*"

The support for a reactor-based source is essentially unanimous in the isotope and activation-analysis community because of the need for the highest possible time-averaged flux. A. Johnson (Oregon State University) stated the case for the activation-analysis group: "... *[the national facility] should incorporate a steady-state source of neutrons and should not be limited to just a pulsed source. Higher power sources will clearly open up new possibilities for an activation of short-lived elements and will provide other new opportunities relative to activation analysis.*" Likewise, the concerns for high-specific-activity isotopes for medical research is voiced by Wynn Volkert (University of Missouri - Columbia): "*Ready accessibility to high thermal neutron flux (i.e., greater than 5×10^{15}) would provide capability to furnish biomedical practitioners and researchers with some important radionuclides that have enough specific activities and large enough quantities that will enormously extend their utility for routine medical applications.*"

Inclusion of the construction costs of most of the instruments is recommended to be part of the ANS overall budget, and the ready availability of these instruments without charge (ideally with facility support for travel and living expenses as at the ILL) to the user community is of equal concern to the construction of the source itself. High-quality versatile instruments are extremely important, as reflected by Bill Yelon (MURR): "*The success of the ILL can be attributed much less to its high flux than to the considerable investments made in modern instruments.*"

5.2.1.2 University "Feeder" Sources

The university community is charged with the responsibility of educating the future generations of neutron scientists and engineers. It is thus suggested that their needs must play an important role in the overall planning and justification for a new national neutron source. The continued health of this segment of the higher education community should be seen as crucial. This health will require *support at the university level*. The building of a new national source in itself will not suffice. It is thus recommended that the budget for a new national source include limited funding to support and maintain regional neutron centers at universities. The amount recommended in the above-referenced NAS study is \$30M (FY 1992 dollars) per year, which is certainly modest on the scale of the ANS, and may represent very cost-effective spending for the future of the field and the ANS. The university support needs to include funds for upgrading existing reactor facilities and instrumentation as well as enhanced staff and operating costs for the facilities. All requests should be based on peer-reviewed specific proposals by the regional centers and judged principally on their applicability to the needs of the new national source. As stated by Myron Salamon, "*While the ANS is certainly a desirable project, I think the neutron scattering community in this country would be better served in the short term by a substantial upgrade of*

existing reactor facilities at Brookhaven, NIST, Oak Ridge, and Missouri. Strong support should be given for significant improvements at IPNS."

5.2.1.3 The Need to Move Expediently and Decisively

The fundamental livelihood of neutron science in the U.S. is on the line. The present sources are of advanced age and badly need modernization as well as replacement within the next decade. The ability to convince students of the viability of a career in neutron sciences and engineering depends on a national decision to support the field *including development of a new reactor source*. The time is at hand to move forward decisively to resuscitate this field and to allow the U.S. to regain its original prominence in this critical area of science. Without such a decision, *"I hesitate to think that I might be a member of the last generation of U.S. neutron scatterers ... meanwhile it continues to be a thriving research area in Europe and Japan,"* as stated by Alan Goldman (Iowa State University).

5.2.2 Reference

1. *University Research Reactors in the United States — Their Role and Value* (National Academy Press, Washington, D.C. 1988).

Appendix A

Advanced Neutron Source Performance Goals and Conceptual Design Parameters

Parameter ^{a,b}	NSCANS Goals	ANS Conceptual Design
<i>Neutron scattering</i>		
Hot neutrons		
Thermal flux at hot source	≥ 1	1
Number of hot sources	1	1
Number of hot beams	2	2
Thermal neutrons		
Peak thermal flux in reflector	5-10	7 ^c
Thermal:fast ratio	$\geq 80:1$	170:1
Number of thermal tangential tubes	7	7
Cold neutrons		
Thermal flux at cold sources	2-4	4
Number of cold sources	2	2
Number of horizontal cold guides	14	14
Number of slant cold beams for scattering instruments	1	1
<i>Nuclear and fundamental physics</i>		
Number of thermal through tubes	1	1
Number of slant thermal beams	1	1
Number of very cold beams	2	2
<i>Materials irradiation^d</i>		
Central irradiation facility		
Fast flux	≥ 1.4	2.0 ^e
Fast:thermal ratio	$\geq 1:2$	2.9:1 ^e
Total number of positions	10	10
Number of instrumented positions	2	5
Damage rate [displacement per atom per year (dpa/yr) in stainless steel]	≥ 30	[TBD] ¹
Nuclear heating rate (W/g in stainless steel)	≤ 54	≤ 70
Axial flux gradient over 200 mm	$\leq 30\%$	14%
Available diameter (mm)	≥ 17	48
Available length (mm)	≥ 500	500

Parameter ^{a,b}	NSCANS Goals	ANS Conceptual Design
<i>Materials irradiation^d (Cont.)</i>		
Reflector vessel facility		
Fast flux	≥ 0.5	0.5
Fast:thermal ratio	$\geq 1:3$	1:13 ^f
Number of instrumented positions	≥ 8	2 ^g
Damage rate (dpa/yr in stainless steel)	≥ 8	[TBD] ¹
Nuclear heating rate (W/g in stainless steel)	≤ 15	18
Axial flux gradient over 200 mm	$\leq 30\%$	16%
Available diameter (mm)	≥ 48	48
Available length (mm)	≥ 500	300 ^h
<i>Isotope production</i>		
Transuranium production		
Epithermal flux	≥ 0.6	1.4
Epithermal:thermal ratio	$\geq 1:4$	1:3
Allowable peak heat flux (MW/m ²)	4	[TBD] ¹
Total annual production:		
²⁵² Cf (g)	1.5	1.7 ⁱ
²⁵⁴ Es (μg)	40	60 ⁱ
Epithermal-hydraulic rabbit tube		
Epithermal flux	flux peak position	1.0 ^j
Epithermal:thermal ratio	$\geq 1:4$	1:4.5 ^j
Allowable peak heat flux (MW/m ²)	1.75	1.75
Other isotope production facilities		
Thermal flux	1	0.9
Number of reflector positions	≥ 4	4
Number of thermal-hydraulic rabbit tubes	3	3
<i>Materials analysis</i>		
Activation analysis pneumatic tubes		
40-cm ³ rabbits in reflector	4	4
2-cm ³ rabbits in reflector	1	1
Thermal flux at reflector rabbit positions	≥ 0.2	0.3 (tube 1) 0.06 (tubes 2, 3, 4, and 5) ^k
Heating rate:		
Temperature in a 40-cm ³ high density polyethylene rabbit (°C)	≤ 120	[TBD] ¹
Rabbit tubes in light-water pool	2	2
Thermal flux at light-water rabbit positions	≥ 0.04	0.05; 0.02

Parameter ^{a,b}	NSCANS Goals	ANS Conceptual Design
<i>Materials analysis (Cont.)</i>		
Prompt-gamma activation-analysis cold neutron stations		
Low-background (multiple-beam) guide system	1	1
Neutron depth profiling		
Number of slant cold beams	1	1
Gamma irradiation facility in spent-fuel pool	≥ 1	1
Positron production position	1	1

^a All fluxes in units of 10^{15} n/cm²·s.

^b Neutron spectra terms as used in this table are defined as follows:

thermal ≤ 0.625 eV 0.625 eV $\leq$ epithermal ≤ 100 eV fast > 0.1 MeV.

Sources of neutrons for research are classified as follows:

ultracold < 25 μ eV 25 μ eV $<$ very cold < 0.1 meV 0.1 meV $<$ cold < 5 meV
 5 meV $<$ thermal < 0.625 eV 0.625 eV $<$ hot < 1 eV.

^c End-of-cycle value.

^d The materials irradiation facilities are intended to replace irradiation facilities in the HFIR. The ANS can not meet all these goals since the simultaneous requirements of high fast:thermal flux-ratio, high-fast flux, and low heating rate are intrinsically incompatible with the physics of an undermoderated core (optimized for neutron scattering research goals).

^e Values at $r = 160$ mm and beginning-of-cycle. The fast flux increases during the fuel cycle. The fast:thermal ratio decreases to as low as 0.25:1 with decreasing radius and increasing burnup.

^f Best available ratio in reflector vessel.

^g Access restricted by other facilities in the reflector vessel. Partially offset by providing more instrumented positions than required in the central irradiation facility.

^h Length restricted by outer shutdown rods.

ⁱ The production number quoted is a preliminary value obtained from a two-dimensional neutronic analysis. Possible thermal limitations on pellet loadings have not been evaluated.

^j Value at 250 mm above midplane at midpoint of cycle.

^k Optimized for lowest gamma heating, but will need to be located at a higher neutron flux level.

^l To be determined.

Appendix B

Current and Projected Isotope Usage in the United States

B.1 Isotopes in Common Usage

Usage	Isotope(s)	No. of Facilities	Use	Percent Research	Percent Clinical
Nuclear Medicine Diagnosis ^a	^{99m} Tc, ¹³¹ I, ¹²⁵ I, ²⁰¹ Tl, ¹⁶⁷ Ga, ¹¹¹ In, ⁶⁷ Cu	Large No.	35,000 procedures/day ^a	1	99
Diagnosis Research ^a	~50 (see Table 4.1)	Large No.	2–3,000 procedures/year ^{a,b}	99	1
Treatment ^a	¹³¹ I	Large No.	10–15,000 procedures/year	0.1	99.9
Radioimmunoassay	¹²⁵ I	Large No.	350,000 procedures/day ^a	8–15	85
Blood Product Irradiation	⁶⁰ Co, ¹³⁷ Cs	Large No.	30–50,000 procedures/day ^a	—	100
Food Irradiation	⁶⁰ Co, ¹³⁷ Cs	100	Large	1	99
Radiation Therapy	⁶⁰ Co, ¹³⁷ Cs, ¹⁹² Ir	Large No.	10–50,000 procedures/day	1	99
Radiation Therapy Research					
Conventional ^a	¹⁰³ Pd, ^c ¹⁴⁵ Sm, ⁸⁹ Sr, ⁶⁷ Cu, ^c ²⁵² Cf, Antibody Therapy ²¹¹ At, ²²⁷ Ac, ²¹³ Bi, ²⁵³ Es, ²⁵⁵ Fm (see Table 4.1)	100	10,000 procedures/year	100	—
Neutron Therapy ^a	²⁵² Cf	10 (estimated 100 in 2001)	Currently, cancer research use only (clinical, physics, radiobiology, radiooncology)	100	—

^a U.S. usage only.

^b Current research on ¹⁴¹Ce and ^{123m}Te for lung density measurements (300–500-mg source; 100 sources/year projected).

^c ¹⁰³Pd, ^{117m}Sn, ⁶⁷Cu — Multigram quantities used annually.

B.2 Current and Projected ^{252}Cf Neutron Usage

	No. of Facilities		Amount Used (mg)	Percent Research	Percent Routine
	Present	Projections ^a			
Power Reactor (PWR)	> 60	60	5-25	—	100
Cancer Therapy	10	100	5-10 ^b	100	—
Airport Luggage Surveillance	8	?	200 ^b	50	50
Biology and Medicine Irradiators and Activation Analysis	4	25	25-100	100	—
Teletherapy	2 ^a	15-25	25-100	100	—
FDA Food (activation analysis)	1	?	200	100	—
Navy (material analysis)	1	?	100	100	—
Industrial Radiography	1	?	100	—	100
Waste Analysis (LANL, TRU, Portsmouth, etc.)	3	?	1	80	20
Cement (on-line activation analysis and monitoring)	Large No.	Large No.	10-500	—	100

^a By year 2001.

^b Total worldwide. Others per user or machine.

B.3 Sales of Reactor-Formed Radioisotopes (IP&D, ORNL)

Isotope	Volume		
	FY 1989	FY 1990	FY 1991
Cobalt 60 (Curies)	1023842	341571	687125
Iridium 192 (Curies)	40876	60586	465838
Nickel 63 (Millicuries)	44676	13464	118000
Tritium (Curies)	671781	666347	497712
Californium 252 (Micrograms)	109253	218487	17647
Gadolinium 153 (Curies)	45		300
Iodine 129 (Milligrams)	3330	2910	
Krypton 85 (Curies)	P 3806	3146	3895
	E 529	549	70
Technetium 99 (Grams)	68	15	255
Curium 244 (Milligrams)	1501	20	
Neptunium 237 (Grams)	3		
Plutonium 238 (Milligrams)	3051	3050	
Plutonium 239 (Milligrams)	60252	1	125
Plutonium 241 (Milligrams)	594		
Plutonium 242 (Milligrams)	10000		100
Uranium 233 (Milligrams)	3210	20106	103
Uranium 236 (Milligrams)	10310	30	3

P – Purified Krypton Gas.

E – Purified Krypton Gas Enriched in ⁸⁵Kr.

Note: Palladium 103 is made at ORNL as a radiation service; the customer provides targets. Irradiated targets are shipped periodically as specified by the customer.

**B.4 Estimate of Future Demand for ⁸⁹Sr
(Joint Action of IP&D, ORNL and Amersham)**

Strontium 89 (Millicuries)	U.S. Market Developing Est. 100,000-1,000,000 yearly
-------------------------------	---

**B.5 Estimated Annual U.S. Markets for Reactor-Formed Radioisotopes
(IP&D, ORNL)**

Isotope	Volume
Molybdenum 99 (6-Day Curies)	104000
Iodine 125 (Curies)	2080
Iodine 131 (Curies)	3200
Xenon 133 (Curies)	27300
Phosphorus 32 (Curies)	2080

Appendix C

Summaries of Reports on Related Uses of Neutron Sources

C.1 Positron Spectroscopy of Materials

A.P. Mills, Jr. (AT&T Bell Laboratories)

Positrons and electrons form complementary probes of condensed matter. The use of electron beams, which are readily available as laboratory tools, is widespread and well understood. Positrons provide a qualitatively different probe. Their positive charge repels them from the interior of materials, for example, so they are effective as a probe of outer electrons in high-Z materials, which are difficult to study with Compton scattering. Positron decay by annihilation with an electron produces two photons. The angular correlation of this annihilation radiation (ACAR) directly reflects the momentum distribution of the annihilating pairs and allows determination of electron momentum distributions. A recent notable example is the measurement of Fermi surfaces in YBCO, Bi-2212, and La₂CuO₄-based high-temperature superconductors; these are among the few available experimental results that can narrow theoretical speculation about these materials.

Positrons and the electron-positron bound state known as positronium are currently used to study electronic structure, atomic physics, molecular physics and chemistry (including liquid and gas phases), defects in metals and alloys, semiconductors, superconductors, and polymers. The negative work function of positrons means that they are, in principle, better surface probes than electrons. Positron and positronium physics and chemistry are research fields in their own right.

Positron research has been inhibited by the difficulty of producing slow positron beams of sufficient intensity. The best positron beam currently available is about 1 nA, compared with about 100 nA in a typical electron microscope. The reactor-produced radioisotopes that yield positron beams (e.g., ⁶⁴Cu, ⁵⁸Co, and ⁷⁹Kr) have high specific activity (i.e., short half-lives) to provide sufficient intensity, so that a high time-averaged neutron flux is essential for their production. The source planned for the ANL will yield e⁺ beams with an intensity (greater than 10 nA from a 0.2-mm source) approaching those found in electron microscopes and will allow positron research to attain its full potential, including new applications such as microprobe analysis and scanning positron microscopy.

C.2 Applications of Muons

S.F.J. Cox (Rutherford Appleton Laboratory, U.K.)

The positive muons, which are produced by accelerator proton beams like those used in a spallation neutron source, provide a powerful nuclear probe for materials and chemical sciences. Implanted in virtually any material, its spin polarization can be monitored through its decay asymmetry to yield information on structural and electronic properties of the host. With its large magnetic moment (three times that of the proton), it is sensitive to local magnetic fields, thus providing data on both the magnitude and the distribution of these fields. Determination of the magnetic penetration depth in superconductors is a well-documented example of the use of this technique. Moreover, both static and dynamic fields can be monitored via the muon relaxation functions and, for example, can give information on magnetic phase transitions. When the μ^+ is implanted in matter, it behaves like a proton, which is especially valuable in situations where hydrogen is difficult or impossible to detect by conventional spectroscopies. For example, virtually all information on isolated hydrogen defect centers in semiconductors comes from studies of the muon analog. Much of our knowledge of quantum diffusion of hydrogen-like defects in metals, semiconductors, and insulators comes from Muon Spin Rotation/Relaxation/Resonance (μ SR). Because a muon production target intercepts little (about 2%) of the proton beams used for neutron production, there is negligible loss of neutron intensity in a "dual-use" facility.

Pulsed muons are also useful for a variety of other fundamental measurements. Beams with pulses of muons significantly shorter than the muon lifetime (2.2 μ s) greatly reduce beam-associated backgrounds, provide a precise zero in time for measurements that study time evolution of a state, facilitate line-narrowing techniques, and allow studies of laser-induced transitions of atoms containing muons. Studies to be pursued include precision measurements of the hyperfine interval and Zeeman effect splitting in muonium (μ^+e^-), which are used to determine the fine structure constant and the ratio of the magnetic moments for the muon and the proton; laser-induced transitions in muonium, which are used to measure the muon mass precisely; and laser spectroscopy measurements in muonic hydrogen and helium, which are used to measure nuclear charge distributions accurately. Other studies include a measurement of the muon lifetime, which provides the best determination of the weak-interaction coupling constant, and muon capture, which is sensitive to hadronic weak couplings. A pulsed-muon facility is also an excellent source of low-energy neutrinos that can be used to study neutrino scattering, which probes higher-order processes in the Standard Model; and neutrino proton scattering, which is an ideal way to determine several weak hadronic coupling constants.

Appendix D

List of Acronyms

The following is a list of acronyms, initialisms, and abbreviations used in this report.

ACAR	angular correlation of annihilation radiation
ANL	Argonne National Laboratory
ANS	Advanced Neutron Source, ORNL
APFIM	atom probe field ion microscopy
APS	Advanced Photon Source, ANL
APT	Accelerator Production of Tritium
ATR	Advanced Test Reactor, INEL
ATW	Accelerator Transformation of Waste
BES	Office of Basic Energy Sciences, DOE
BESAC	Basic Energy Sciences Advisory Committee
BLIP	Brookhaven Linear Isotope Producer, BNL
BNL	Brookhaven National Laboratory
CCD	charge-coupled device
CEBAF	Continuous Electron Beam Accelerator Facility, Newport News, Virginia
CERN	Centre Europeenne de Recherches Nucleaires, Switzerland
CID	charge-injected device
CNRF	Cold Neutron Research Facility, NIST
CRNL	Chalk River Nuclear Laboratories, Canada
CW	constant wavelength
DOC	U.S. Department of Commerce

DOE	U.S. Department of Energy
dpa	displacements per atom
DRAM	dynamic random access memory
EBR-II	Experimental Breeder Reactor II, ANL
EDM	electron dipole moment
ES&H	environment, safety, and health
ESS	European Spallation Source
FFTF	Fast Flux Test Facility, Hanford, Washington
FRM	Reactor, Germany
FY	fiscal year
GDP	Gross Domestic Product
GRID	gamma-ray-induced Doppler broadening
GSI	Gesellschaft für Schwerionen Physik, Darmstadt, Germany
HFBR	High-Flux Beam Reactor, BNL
HFIR	High-Flux Isotope Reactor, ORNL
HFR	High-Flux Reactor, ILL
HMI	Hahn-Meitner Institute, Berlin, Germany
ILL	Institut Laue-Langevin, Grenoble, France
INEL	Idaho National Engineering Laboratory
INS	inelastic neutron scattering
IPNS	Intense Pulsed Neutron Source, ANL
IP&D	Isotope Production and Distribution, ORNL
ISIS	Pulsed Spallation Source, RAL, U.K.

JAERI	Japan Atomic Energy Research Institute, Tokai-mura
JRR	Japan Research Reactor, Tokai-mura
KENS	KEK (Japan High-Energy Physics Laboratory) Neutron Source, Tsukuba
KFA	Kernforschungsanlage, Jülich, Germany
LAMPF	Los Alamos Meson Physics Facility, LANL
LANL	Los Alamos National Laboratory
LANSC	Los Alamos Neutron Scattering Center, LANL
LASREF	Los Alamos Spallation Radiation Effects Facility, LANL
LIA	linear induction accelerator
LBL	Lawrence Berkeley Laboratory
LWR	light water reactor
L/D	length-to-diameter ratio
MSANS	multiple small-angle neutron scattering
MURR	Missouri University Research Reactor, Columbia, Missouri
NAS	U.S. National Academy of Sciences
NBSR	Neutron Beam Split-Core Reactor, NIST
NIST	National Institute of Standards and Technology
NMR	nuclear magnetic resonance
NRC	U.S. Nuclear Regulatory Commission
NSCANS	U.S. National Steering Committee for the Advanced Neutron Source
NSE	neutron spin echo
NSLS	National Synchrotron Light Source, BNL
NSSA	Neutron Scattering Society of America

OER	Office of Energy Research, DOE
ORELA	Oak Ridge Electron Linear Accelerator, ORNL
ORNL	Oak Ridge National Laboratory
ORPHEE	Reactor, Saclay, France
PET	positron emission tomography
PSD	positron-sensitive detector
PSI	Paul Scherrer Institute, Switzerland
PSNS	pulsed spallation neutron source
QA	quality assurance
RAL	Rutherford Appleton Laboratory, Chilton, U.K.
RCS	rapid-cycling synchrotron
REDC	Radiochemical Engineering Development Center, ORNL
RERTR	Reduced Enrichment Research and Testing Reactor, ANL
RFQ	radio-frequency quadropole
R&D	research and development
SANS	small-angle neutron scattering
SATURNE	Accelerator, Saclay, France
SIN	Schweizerisches Institute für Nuklearforschung, Villeggen, Switzerland (former name for the Paul Scherrer Institute)
SINQ	Schweizerisches Institute für Nuklearforschung Quelle (Swiss Institute of Nuclear Research Source), Villeggen, Switzerland
SNQ	Spallations Neutronen Quelle (Spallation Neutron Source), Germany
TAS	triple-axis spectroscopy
TEC	total estimated cost

TEM	transmission electron microscopy
TOF	time-of-flight
TRIUMF	Tri-University Meson Facility, Canada
UCN	ultracold neutron
VLSI	very-large-scale integration
WWR-M	Reactor, St. Petersburg Nuclear Physics Institute, Russia
μSR	Muon Spin Rotation/Relaxation/Resonance
2-D	two dimensional
3-D	three dimensional

DATE

FILMED

3/10/94

END